DIELECTRIC ELASTOMER COMPOSITES: ANALYTICAL AND NUMERICAL NON-CONVEX HOMOGENIZATION METHODS AND APPLICATIONS

BY

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DISSEYATION

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ABSTRACT

DIELECTRIC ELASTOMER COMPOSITES: ANALYTICAL AND NUMERICAL
NON-CONVEX HOMOGENIZATION METHODS AND APPLICATIONS

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With the practical objective of shedding light on promising experimental results that have recently identified dielectric elastomer composites as potential enablers of new technologies (essentially, as the next generation of soft sensors and actuators), this work puts forth analytical and numerical methods to determine the macroscopic elastic dielectric behavior of this class of soft electroactive materials directly in terms of their microscopic behavior.

The macroscopic behavior of dielectric elastomer composites is first investigated within the classical asymptotic context of small deformations and moderate electric fields. Specifically, by a combination of analytical and numerical techniques, rigorous homogenization solutions are constructed for dielectric elastomer composites with general (possibly anisotropic) classes of two-phase particulate microstructures. Aimed at identifying what types of filler particles lead to enhanced elastic dielectric behaviors, these solutions are deployed to examine dielectric elastomers filled with stiff high-permittivity particles, high-permittivity particles that are liquid-like in mechanical behavior, and vacuous pores.

In addition to generalizing the fundamental purely elastic and purely dielectric solutions of Es-helby and Maxwell to the coupled and nonlinear realm of electroelastostatics, the above-outlined rigorous asymptotic solutions turn out to be essential in the development of corresponding homogenization solutions for finite deformations and finite electric fields. Indeed, it is shown that they can be utilized as building blocks for the derivation of a general approximate homogenization solution for non-Gaussian dielectric elastomers filled with nonlinear elastic dielectric particles that may exhibit polarization saturation. By construction, this approximate solution is exact in the limit of small deformations and moderate electric fields. For finite deformations and finite electric fields, its accuracy is assessed by direct comparisons with full-field hybrid finite-element simulations, as well as with numerical solutions generated via a new WENO finite-difference scheme developed specifically for this class of problems. With the object of scrutinizing recent experimental results, the
specializations of the proposed solution to various cases wherein the filler particles are of poly- and mono-disperse sizes and exhibit different types of elastic dielectric behaviors are discussed in detail.

Stark disagreement between the theoretical results outlined above and a plurality of experimental results indicates that the basic point of view that dielectric elastomer composites can be idealized as two-phase particulate elastic dielectric composites is fundamentally incomplete, especially for cases involving stiff filler particles which (as opposed to what the theory predicts) have been reported to exhibit extreme enhancements in their electrostriction capabilities. It is posited that such extreme enhancements are the manifestation of interphasial phenomena. In particular, the presence of interphasial free charges that oscillate rapidly in space at the length scale of the microstructure of elastic dielectric composites is shown to have a significant and even dominant effect on their macroscopic response, possibly leading to extreme behaviors ranging from unusually large permittivities and electrostriction coefficients to metamaterial-type properties featuring negative permittivities. These results suggest a promising strategy to design deformable dielectric composites — such as electrets and dielectric elastomer composites — with exceptional electromechanical properties.
A ma famille
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Introduction

Running, one might say, is basically an absurd past-time upon which to be exhausting ourselves. But if you can find meaning in the kind of running you have to do to stay on this team, chances are you will be able to find meaning in another absurd past-time: Life.

— Bill Bowerman (Donald Sutherland), Without Limits, Dir. Robert Towne, 1998

Deformable dielectrics have repeatedly been called upon to enable new technologies. This has been true again since the turn of the millennium when material scientists “rediscovered” soft organic dielectrics — the core subject of this thesis — as a class of materials with the potential to enable the next generation of soft sensors and actuators.

The history of deformable dielectrics is a motley one and worth recalling here before we zero in on soft organic dielectrics. The electromechanical coupling at play in such electroactive materials was originally unveiled by the brothers Pierre and Jacques Curie at the end of the nineteenth century. They discovered (Curie and Curie, 1880) that positive and negative electric charges would appear on parts of the surface of some naturally occurring crystals such as quartz, tourmaline, and Rochelle salt\(^1\), when mechanically compressed. Following the subsequent theoretical predictions of Lippmann (1881), Pierre and Jacques Curie provided experimental evidence of the existence of a converse effect (Curie and Curie, 1881), namely, that an electric field externally applied on the same materials would induce mechanical deformations. In these sets of experiments, the amount of surface charges induced by external pressures, as well as the deformations induced by external electric fields were linearly related to one another. This *odd* coupling between mechanical and electric fields

\(^{1}\)Rochelle salt or potassium sodium tartrate tetrahydrate was first synthesized in the 1670s by Pierre Seignette, an apothecary native of the coastal town of La Rochelle, France, hometown of the author.
led to the theory of piezoelectricity\textsuperscript{2} and the experimental study of the piezoelectric properties of first a variety of naturally occurring crystals (see, e.g., the historical review of Cady, 1946), and later the now popular synthetic polycrystalline ceramics such as barium titanate (BaTiO\textsubscript{3}) and lead zirconate titanate (Pb\(\left[\text{Zr}_x\text{Ti}_{1-x}\right]\)O\textsubscript{3} \(0 \leq x \leq 1\)) (see, e.g., the monograph of Jaffe et al., 1971). Odd electromechanical coupling has also been observed in electrets (see, e.g., the monograph of Sessler and Gerhard-Multhaupt, 1998), and in certain polymers such as poly(vinylidene fluoride) (PVDF) and copolymers of vinylidene fluoride (see, e.g., the review of Lovinger, 1983). As opposed to the above-described odd coupling, even electromechanical coupling is present in all dielectrics. While the effects of this even coupling is in general negligible in “hard” crystalline and glassy materials, they are dominant in soft organic materials. Notable examples include the electron-irradiated poly(vinylidene-fluoride-trifluoroethylene) copolymer (P(VDF-TrFE)), polyurethane (PU), silicone (PDMS), and acrylic elastomers (see, e.g., Zhang et al., 1998; Pelrine et al., 1998; Pelrine et al., 2000).

The successful career of deformable dielectrics as enablers of new technologies started about four decades after their scientific discovery. Paul Langevin, a former student of Pierre Curie, famously employed for the first time in 1917 piezoelectric crystals as emitters and receptors of ultrasonic waves for submarine sonar detectors. Since then, hard deformable dielectrics with odd electromechanical coupling have enabled a diverse array of technologies, including piezoelectric microphones, loudspeakers, ultrasonic transducers, energy-harvesting devices, resonators in electronic clocks, watches, and radars (see, e.g., Uchino, 1997; Erturk and Inman, 2011; Zelenka, 1986). As already alluded to above, since the turn of the millennium, a wide range of emerging technologies have also been envisioned for dielectric elastomers featuring even electromechanical coupling. Examples include biologically inspired robotics, medical implants, and energy harvesting devices among many others (see, e.g., Bar-Cohen, 2001; Carpi and Smela, 2009; Kornbluh et al., 2011). Despite the impetus provided by this engineering vision, the actual use of dielectric elastomers in new technologies has remained limited, primarily because of the very large electric fields (> 100 MV/m) that they require to achieve meaningful deformations.

Now, as is the case for hard deformable dielectrics with odd electromechanical coupling, it has been recently recognized that the electromechanical properties of dielectric elastomer composites

\textsuperscript{2}The word piezoelectricity derives from the Ancient Greek verb πιέζειν (piezein) meaning “to press” and the noun ἤλεκτρον (electron) for amber. Ancient Greeks had already noticed that amber could attract light particles after being rubbed with fur.
1. Introduction

(sometimes abbreviated as DECs in the rest of this document) outperform those of homogeneous or single-phase dielectric elastomers. Specifically, particulate dielectric elastomer composites, comprising a dielectric elastomer filled with high-permittivity or (semi-)conducting particles, have demonstrated potential to circumvent the shortcomings that have hindered the exploitation of homogenous dielectric elastomers in applications. As a representative example that will be revisited later, we mention here the work of Huang et al. (2005) who have reported a twenty fold enhancement in the uniaxial electrostriction (to be defined in precise terms below) of a PU elastomer filled with semi-conducting o-CuPc particles.

As a clarification, we note that throughout this dissertation composite materials refer to materials with statistically uniform (i.e., translation invariant) heterogeneous dielectric properties. The characteristic length scale at which underlying microstructural traits vary is assumed to be much smaller than the characteristic length scale of the composite material itself. This corresponds in the present context to microscopic length scales roughly between 10 nm and 100 µm. Furthermore, these length scales are sufficiently larger than the atomic/molecular length scale (≈ 10 nm) in order to consider and model these materials as a continuum.

To put the work presented in this dissertation in perspective, it is fitting to recall that conservative\(^3\) macroscopic (or phenomenological) theories of deformable dielectrics were formulated in the first half of the twentieth century by Voigt (1910) for linear piezoelectric materials and later by Toupin (1956) for finitely deformable dielectrics with nonlinear electroelastic couplings. Motivated by the renewed interest in dielectric elastomers, more convenient yet equivalent formulations of Toupin’s theory of elastic dielectrics have been proposed by Dorfmann and Ogden (2005a), McMeeking and Landis (2005), Fosdick and Tang (2007), Vu and Steinmann (2007), Suo et al. (2008), Xiao and Bhattacharya (2008), amongst others. By contrast, microscopic (or homogenization) theories required to characterize the macroscopic elastic dielectric behavior of elastic dielectric composites are sparser. Of course, there are results for linear composites comprising piezoelectric materials (see, e.g., Milton, 2002 and references therein; Spinelli and Lopez-Pamies, 2014). Formulating analogous theories for dielectric elastomer composites with even electroelastic coupling amounts to solving a nonlinear homogenization problem, even within the classical limit of small deformations and moderate electric fields (see, e.g., Section 2.25 in Stratton, 1941; Toupin, 1956).

\(^3\)That is, theories that neglect dielectric and mechanical dissipation.
In the asymptotic context of small deformations and moderate electric fields, heuristic approximations for nonlinear dielectric elastomer composites have been proposed by Li and Rao (2004), Li et al. (2004), and more recently by Siboni and Ponte Castañeda (2013) for the specific case when the particles are mechanically rigid, by making use of classical linear estimates of the Hashin-Shtrikman and self-consistent type. It was in a later contribution that Tian (2007) and Tian et al. (2012) established rigorously via a two-scale convergence argument the homogenization limit of the equations of dielectric elastomer composites in this asymptotic context.

In the general context of finite deformations and finite electric fields, analytical solutions were obtained by deBotton et al. (2007) for the special case of two-phase laminates, Ponte Castañeda and Siboni (2012) later proposed a decoupling approximation for the modeling of a special class of elastic dielectric composites filled with mechanically rigid particles, and Lopez-Pamies (2014) just recently put forth a constitutive theory for a wide class of two-phase particulate microstructures. From a computational perspective, non-hybrid finite-element (FE) formulations for dielectric elastomer composites appear to have been first reported by Li and Landis (2012) and by Keip et al. (2014) in the context of two spatial dimensions, while non-hybrid FE formulations in three dimensions have recently been reported by Miehe et al. (2016); see also the work of Pelteret et al. (2016) on quasi-incompressible media immersed in free space.

The work presented in this dissertation aims at providing a descriptive and predictive microscopic continuum theory for the elastic dielectric behavior of dielectric elastomer composites directly in terms of their underlying microstructure and the constitutive behaviors of their constituents. For relative simplicity, irreversible (or dissipative) mechanical and/or dielectric phenomena such as viscoelasticity, fracture, dielectric loss, electric breakdown are not accounted for in the work presented in this document.

This dissertation is divided into chapters with content extracted from journal articles that have already been published, are under review, or are in preparation to be submitted for publication. For convenience, we detail next the content of each chapter and list for reference purposes the articles they relate to at the end of this introduction.

The next chapter (Chapter 2) consists of the mathematical formulation of the primary electro- elastostatic problem of interest in this dissertation, namely, the characterization of the macroscopic elastic dielectric behavior of dielectric elastomer composites in terms of their microscopic behavior in the absence of space charges. The more general formulation of macroscopic elastic dielectric behavior
in the presence of space charges is deferred to Chapter 9. For arbitrary large deformations and electric fields, this problem takes the form of a nonlinear coupled non-convex variational principle over two sets of (mechanical and electric) variables. Its specialization to the classical asymptotic limit of small deformations and moderate electric fields is subsequently recorded for later use in Chapters 3 through 6. In this limit, the overall elastic dielectric response is characterized by three effective tensors: a fourth-order tensor describing the elasticity of the material, a second-order tensor describing its permittivity, and a fourth-order tensor describing its electrostrictive response. Remarkably, in spite of the inherent coupling and nonlinearity of the problem, Tian et al. (2012) have proved that it is possible to write formulae for the three effective electromechanical tensors characterizing the overall response of dielectric elastomer composites in this asymptotic context in terms of solutions of a system of two uncoupled linear partial differential equations.

Chapter 3, which corresponds to references 2 and 3 in the list of publications, presents a solution for the three effective tensors characterizing the electromechanical response of a general class of two-phase dielectric elastomer composites with (random or periodic) particulate microstructures in the classical limit of small deformations and moderate electric fields. Closed-form formulae are derived for the elastic, dielectric, and electrostrictive effective tensors directly in terms of the corresponding tensors describing in this limit the electromechanical response of the underlying matrix and fillers, and the one- and two-point correlation functions describing the microstructure. This is accomplished by specializing the iterative homogenization theory in finite electroelastostatics of Lopez-Pamies (2014) to the case of elastic dielectrics with even electromechanical and, subsequently, carrying out the pertinent asymptotic analysis. Explicit formulae are recorded for the specific case when the underlying infinitely polydisperse iterative microstructure exhibits the same one- and two-point correlation functions as a transversely isotropic distribution of aligned spheroidal particles. We spell out the five (two elastic, one dielectric, and two electrostrictive) independent effective constants defining the overall elastic dielectric response for the practical case of elastomers filled with an isotropic distribution of spherical particles. Analogous expressions are also provided for the thirteen (five elastic, two dielectric, and six electrostrictive) effective constants defining the overall elastic dielectric response of elastomers filled with a transversely isotropic distribution of aligned cylindrical fibers with circular cross section.

Chapter 4, which corresponds to references 1 and 3, presents solutions derived for the homogenization problem of the elastic dielectric response — still in the limit of small deformations and
moderate electric fields — of an elastomeric matrix comprising differentially coated microstructures. This choice of microstructures allows for an analytical specialization of the theory proposed by Tian et al. (2012). Specifically, results are obtained for isotropic distributions of spherical particles of polydisperse sizes and for transversely isotropic distributions of aligned polydisperse cylindrical fibers with circular cross section. Analytical formulae at hand are obtained for the five and thirteen effective constants defining the macroscopic electromechanical behavior of the corresponding dielectric elastomer composites.

Chapter 5, which corresponds to references 2 and 3, presents a hybrid FE framework employed to obtain the electromechanical response of dielectric elastomer composites in the limit of small deformations and moderate electric fields. This is achieved by constructing numerical solutions for the two partial differential equations entering the theory of Tian et al. (2012), from which, as in Chapter 4, the effective electromechanical properties of the underlying composites can be obtained. Without loss of generality, this framework is presented for composites comprising isotropic distributions of spherical particles of monodisperse size and then recast as a two-dimensional problem for composites comprising transversely isotropic distributions of aligned monodisperse cylindrical fibers with circular cross section.

Chapter 6, which corresponds to references 1, 2, and 3 presents sample results for the effective electromechanical behavior of dielectric elastomer composites in the limit of small deformations and moderate electric fields. The analytical formulas presented in Chapters 3, 4 are compared with corresponding numerical solutions obtained with the hybrid FE framework presented in Chapter 5. These formulae are shown to also be applicable to isotropic suspensions of monodisperse spherical particles or transversely isotropic suspensions of monodisperse aligned fibers with circular cross section, provided that the filler concentration is sufficiently away from percolation. Additionally, with the aim of gaining physical insight into these solutions and shedding light on recently reported experiments, specific results are examined for the practically relevant case of dielectric elastomers filled with isotropic distributions of spherical particles with various elastic dielectric properties, including stiff high-permittivity particles, liquid-like high-permittivity particles, and vacuous pores. Analogous results for transversely isotropic distributions of aligned cylindrical fibers with circular cross section are also recorded.

Chapter 7, which corresponds to references 4 and 8, puts forth homogenization solutions for the macroscopic elastic dielectric response — under finite deformations and finite electric fields — of
ideal elastic dielectric composites with two-phase isotropic particulate microstructures. Specifically, solutions are presented for three classes of microstructures: i) an isotropic iterative microstructure wherein the particles are infinitely polydisperse in size, ii) an isotropic distribution of polydisperse spherical particles of a finite number of different sizes, and iii) an isotropic distribution of monodisperse spherical particles. The solution for the iterative microstructure, which corresponds to the viscosity solution of a Hamilton-Jacobi equation in five “space” variables, is constructed by means of a novel high-order WENO finite-difference scheme. On the other hand, the solutions for the microstructures with spherical particles are constructed by means of hybrid finite elements. Prompted by the functional features shared by all three obtained solutions, a simple closed-form approximation is proposed for the macroscopic elastic dielectric response of ideal elastic dielectric composites with any type of (non-percolative) isotropic particulate microstructure.

Chapter 8, which corresponds to reference 5, presents an analytical framework to construct approximate homogenization solutions for the macroscopic elastic dielectric response — under finite deformations and finite electric fields — of dielectric elastomer composites with two-phase isotropic particulate microstructures. The central idea consists in employing the homogenization solution derived in Chapter 7 for ideal elastic dielectric composites within the context of a nonlinear comparison medium method to generate, in turn, a corresponding solution for composite materials with non-ideal elastic dielectric constituents. Complementary to this analytical framework, a hybrid FE formulation to construct homogenization solutions numerically in three dimensions is also presented. The proposed analytical framework is utilized to work out a general approximate homogenization solution for non-Gaussian dielectric elastomers filled with nonlinear elastic dielectric particles that may exhibit polarization saturation. Exact in the limit of small deformations and moderate electric fields by construction, the solution is shown to accurately apply to arbitrary (non-percolative) isotropic distributions of filler particles for finite deformations and finite electric fields, by means of direct comparisons with FE solutions. Aimed at gaining physical insight into the extreme enhancement in electrostriction properties displayed by emerging dielectric elastomer composites, various cases wherein the filler particles are of poly- and mono-disperse sizes and exhibit different types of elastic dielectric behavior are discussed in detail. Contrary to an initial conjecture in the literature, it is found (inter alia) that the isotropic addition of a small volume fraction of stiff (semi-)conducting/high-permittivity particles to dielectric elastomers does not lead to the extreme electrostriction enhancements observed in experiments. It is posited that such extreme
enhancements are the manifestation of interphasial phenomena.

In support of the conjecture put forth in Chapter 8, Chapter 9, which corresponds to reference 6, presents the derivation of the homogenized equations for the macroscopic response of elastic dielectric composites containing space charges (i.e., electric source terms) that oscillate rapidly in space at the length scale of the microstructure. The derivation is carried out in the setting of small deformations and moderate electric fields by means of a two-scale asymptotic analysis. Two types of rapidly oscillating space charges are considered: passive and active. The latter type corresponds to space charges that appear within the composite in response to externally applied electrical stimuli, while the former corresponds to space charges that are present within the composite from the outset. The obtained homogenized equations reveal that the presence of (passive or active) space charges within elastic dielectric composites can have a significant and even dominant effect on their macroscopic response, possibly leading to extreme behaviors ranging from unusually large permittivities and electrostriction coefficients to metamaterial-type properties featuring negative permittivities. These results suggest a promising strategy to design deformable dielectric composites — such as electrets and dielectric elastomer composites — with exceptional electromechanical properties.

Chapter 10, which corresponds to reference 7, leverages and extends from $N = 3$ to 2 space dimensions the results presented in Chapters 7 and 8 in order to put forth, within the mathematically analogous setting of magnetoelastostatics, an approximate analytical solution for the effective free-energy function describing the homogenized (or macroscopic) magnetoelastic response of magnetorheological elastomers comprised of non-Gaussian rubbers filled with isotropic suspensions of either iron or ferrofluid particles. The solution is general in that it is valid for $N = 2$ and 3 space dimensions and any arbitrary (non-percolative) isotropic suspension of filler particles. By construction, it is exact in the limit of small deformations and moderate magnetic fields. For finite deformations and finite magnetic fields, its accuracy is demonstrated by means of direct comparisons with full-field simulations for two prominent cases for which the specialization of the solution is worked out and discussed in detail: (i) isotropic suspensions of circular particles and (ii) isotropic suspensions of spherical particles. With the combined objectives of demonstrating the possible benefits of using ferrofluid particles in lieu of the more conventional iron particles as fillers and gaining insight into recent experimental results, the proposed homogenization-based constitutive model is deployed to generate numerical solutions for boundary-value problems of both fundamental and practical significance: those consisting of magnetorheological elastomer specimens of spherical and cylindrical shape.
that are immersed in air and subjected to a remotely applied uniform magnetic field. It is found that magnetorheological elastomers filled with ferrofluid particles can exhibit magnetostrictive capabilities far superior to those of magnetorheological elastomers filled with iron particles. The results also reveal that the deformation and magnetic fields are highly heterogenous within the specimens and strongly dependent on the shape of these, specially for magnetorheological elastomers filled with iron particles. From an applications perspective, this evidence makes it plain that attempts at designing magnetostrictive devices based on magnetorheological elastomers need to be approached, in general, as structural problems, and not simply as materials design problems.

Finally, Chapter 11 records some concluding remarks as well as prospects for future work.
List of publications that have resulted from this dissertation work


2.

The problem

Mathematics, rightly viewed, possesses not only truth, but supreme beauty—a beauty cold and austere, like that of sculpture, without appeal to any part of our weaker nature, without the gorgeous trappings of painting or music, yet sublimely pure, and capable of a stern perfection such as only the greatest art can show.

– Bertrand Russell, *A History of Western Philosophy*, 1945

This chapter is concerned with the mathematical formulation of the homogenization problem in finite electroelastostatics that describes the macroscopic elastic dielectric behavior of dielectric elastomer composites in the absence of space charges. We begin in Section 2.1 by introducing the microscopic description of dielectric elastomer composites. In Section 2.2, we formulate the homogenization problem that defines their macroscopic elastic dielectric behavior for arbitrarily large deformations and electric fields. For later use in Chapters 3 through 6, we spell out in Section 2.2.1 the specialization of such a macroscopic behavior in the classical asymptotic limit of small deformations and moderate electric fields.

2.1 Microscopic description of dielectric elastomer composites

Consider a dielectric elastomer composite that, in its undeformed configuration, occupies the domain $\Omega \subset \mathbb{R}^3$ with boundary $\partial \Omega$. For convenience, we choose units of length so that $\Omega$ has unit volume. Each material point in $\Omega$ is identified by its position vector $\mathbf{X}$, while its position in the deformed
configuration $\Omega_d \subset \mathbb{R}^3$ is given by
\[ x = \chi(X). \quad (2.1) \]

In order to satisfy the physical requirement of material impenetrability, $\chi$ is assumed to be a one-to-one mapping on $\Omega$, that is,
\[ \chi(X) = \chi(X') \iff X = X', \quad X, X' \in \Omega. \quad (2.2) \]

Furthermore, $\chi$ is assumed to be twice continuously differentiable, except possibly on surfaces of discontinuity of material properties, where it is only required to be continuous. The corresponding deformation gradient at $X$ is denoted by $F$ and given by
\[ F = \frac{\partial \chi}{\partial X}(X). \quad (2.3) \]

From (2.2) and continuity arguments it follows that
\[ \det F > 0, \quad X \in \Omega. \quad (2.4) \]

The constitutive behavior of the dielectric elastomer composite is taken to be characterized by a “total” free energy $W$ per unit undeformed volume that is a non-convex objective function of the deformation gradient tensor $F$ and an objective function of the Lagrangian electric field $E$:
\[ W = W(X, F, E) = W(X, QF, E) \text{ for all orthogonal second-order tensors } Q \in \text{Orth}^+ \text{ and all } F \text{ and } E. \]

Note that the Eulerian electric field $e$ is given by
\[ e = F^{-T}E. \quad (2.5) \]

Furthermore, motivated by experimental evidence, this total free energy is taken to be an even function of $E$, that is, $W(X, F, E) = W(X, F, -E)$. It follows that at each material point $X \in \Omega$ the first Piola-Kirchhoff stress tensor $S$ and the Lagrangian electric displacement field $D$ are given in terms of $F$ and $E$ simply by (see, e.g., Dorfmann and Ogden, 2005a)
\[ S = \frac{\partial W}{\partial F}(X, F, E) \quad \text{ and } \quad D = -\frac{\partial W}{\partial E}(X, F, E). \quad (2.6) \]

The Cauchy stress $T$, Eulerian electric displacement $d$, and polarization $p$ (per unit deformed volume) are in turn given by
\[ T = \frac{1}{\det F}SF^T, \quad d = \frac{1}{\det F}FD, \quad \text{and} \quad p = d - \varepsilon_0 F^{-T}E, \quad (2.7) \]

\footnote{In this context, the adjective “total” means that the free energy density for an elastic dielectric material has been suitably amended to account for the effects of the so-called Maxwell stress (see, e.g., Dorfmann and Ogden, 2005a).}
where $\varepsilon_0 \approx 8.85 \times 10^{-12}$ F/m stands for the permittivity of vacuum.

In the context of electroelastostatics (see, e.g., Kovetz, 2000), balance of momenta and Maxwell’s equations require that

$$\text{Div} \mathbf{S} = 0, \quad \mathbf{S} \mathbf{F}^T = \mathbf{F} \mathbf{S}^T, \quad \text{Div} \mathbf{D} = 0, \quad \text{Curl} \mathbf{E} = 0, \quad \mathbf{X} \in \Omega,$$

(2.9)

where we have assumed the absence of body forces, space charges, free currents, and magnetic fields. It follows from the assumed objectivity of the free energy $W$ that the balance of angular momentum (2.9) is automatically satisfied. It also follows from Faraday’s law (2.9) that

$$\mathbf{E} = -\text{Grad} \Phi,$$

(2.10)

where the scalar field $\Phi$ has been introduced to denote the electric potential. The only non-trivial equations that remain from (2.9) are thus the balance of linear momentum and Gauss’s law:

$$\text{Div} \mathbf{S} = 0 \quad \text{and} \quad \text{Div} \mathbf{D} = 0, \quad \mathbf{X} \in \Omega.$$

(2.11)

At this point, we note that at several passages in this document it will prove useful to treat the Lagrangian electric displacement field $\mathbf{D}$ as the independent electric variable instead of the Lagrangian electric field $\mathbf{E}$. To avoid loss of continuity, details for such a formulation are deferred to Appendix A.

**Two-phase particulate dielectric elastomer composites.** In most of this dissertation, attention is restricted to the fundamental case of two-phase particulate dielectric elastomer composites. A schematic is illustrated in Fig. 2.1. Specifically, we consider dielectric elastomer composites made out of a homogeneous elastic dielectric matrix, which occupies a continuous domain $\Omega^{(1)} \subseteq \Omega$, filled with a statistically uniform distribution of homogeneous elastic dielectric particles occupying disconnected domains. The union of these disconnected domains is denoted by $\Omega^{(2)}$, so that $\Omega = \Omega^{(1)} \cup \Omega^{(2)}$ and $\Omega^{(1)} \cap \Omega^{(2)} = \emptyset$. Consistent with our definition of composite materials, the characteristic length scale of the filler particles is taken to be much smaller than the size of $\Omega$. The domains that they

---

2At surfaces of material discontinuity $\Gamma$ with normal vector $\mathbf{N}$, equations (2.9) should be interpreted as the jump conditions

$$[\mathbf{S}] \mathbf{N} = 0, \quad [\mathbf{D}] \cdot \mathbf{N} = 0, \quad [\mathbf{E}] \times \mathbf{N} = 0, \quad \mathbf{X} \in \Gamma.$$

(2.8)

3The presence of space charges will be considered in Chapter 9.
occupy can be conveniently described with help of the characteristic or indicator function

\[
\theta(\mathbf{X}) = \begin{cases} 
1 & \text{if } \mathbf{X} \in \Omega^{(2)} \\
0 & \text{otherwise}
\end{cases}
\]  

(2.12)

Their distribution may be periodic or random. For periodic distributions of filler particles, the characteristic function \( \theta \) is deterministically known once a unit cell and the lattice over which it is repeated are specified. For random distributions, the indicator function \( \theta \) is known only in a probabilistic sense.

Since both the matrix (\( r = 1 \)) and the filler particles (\( r = 2 \)) are homogeneous elastic dielectrics, we write

\[
W(\mathbf{X}, \mathbf{F}, \mathbf{E}) = [1 - \theta(\mathbf{X})]W^{(1)}(\mathbf{F}, \mathbf{E}) + \theta(\mathbf{X})W^{(2)}(\mathbf{F}, \mathbf{E}),
\]

(2.13)

where \( W^{(1)} \) stands for the free-energy function describing the behavior of the matrix, while \( W^{(2)} \) stands for the free-energy function describing the behavior of the fillers. Note that \( W^{(r)}(\mathbf{QF}, \mathbf{E}) = W^{(r)}(\mathbf{F}, \mathbf{E}) = W^{(r)}(\mathbf{F}, -\mathbf{E}) \) for all \( \mathbf{Q} \in \text{Orth}^+, \) all \( \mathbf{F} \) and \( \mathbf{E} \), and \( r = 1, 2 \).

Figure 2.1: Microscopic view of a two-phase particulate dielectric elastomer composite.

2.2 The macroscopic response

Granted the separation of length scales and statistical uniformity of the microstructure, the above-defined dielectric elastomer composite behaves macroscopically as a “homogenous” material. Its
macroscopic response is defined by the relation between the volume averages of the first Piola-
Kirchhoff stress tensor and Lagrangian electric displacement,

\[ \mathbf{\bar{S}} = \int_{\Omega} \mathbf{S}(\mathbf{X}) d\mathbf{X} \quad \text{and} \quad \mathbf{\bar{D}} = \int_{\Omega} \mathbf{D}(\mathbf{X}) d\mathbf{X}, \]  

(2.14)

and the volume averages of the deformation gradient \( \int_{\Omega} \mathbf{F}(\mathbf{X}) d\mathbf{X} \) and Lagrangian electric field \( \int_{\Omega} \mathbf{E}(\mathbf{X}) d\mathbf{X} \) over the undeformed configuration \( \Omega \) when the composite is subjected to affine boundary conditions (Hill, 1972). Consistent with our choice of \( \mathbf{F} \) and \( \mathbf{E} \) as the independent variables, we consider the case of affine deformation and affine electric potential:

\[ \mathbf{x} = \mathbf{F} \mathbf{X} \quad \text{and} \quad \Phi = -\mathbf{E} \cdot \mathbf{X}, \quad \mathbf{X} \in \partial \Omega, \]  

(2.15)

where the second-order tensor \( \mathbf{F} \) and vector \( \mathbf{E} \) stand for prescribed boundary data. It follows from the divergence theorem that

\[ \mathbf{F} = \int_{\Omega} \mathbf{F}(\mathbf{X}) d\mathbf{X} \quad \text{and} \quad \mathbf{E} = \int_{\Omega} \mathbf{E}(\mathbf{X}) d\mathbf{X}. \]  

(2.16)

The sought macroscopic constitutive relation between \( \mathbf{S}, \mathbf{D} \) and \( \mathbf{F}, \mathbf{E} \) can be compactly written as (Lopez-Pamies, 2014)

\[ \mathbf{S} = \frac{\partial W}{\partial \mathbf{F}} (\mathbf{F}, \mathbf{E}, c) \quad \text{and} \quad \mathbf{D} = -\frac{\partial W}{\partial \mathbf{E}} (\mathbf{F}, \mathbf{E}, c), \]  

(2.17)

where

\[ \overline{W} (\mathbf{F}, \mathbf{E}, c) = \min_{\mathbf{F} \in \mathcal{K}} \max_{\mathbf{E} \in \mathcal{E}} \int_{\Omega} W(\mathbf{X}, \mathbf{F}, \mathbf{E}) d\mathbf{X}, \]  

(2.18)

the effective free energy function, corresponds physically to the total electroelastic free energy (per unit undeformed volume) of the dielectric elastomer composite. In these last expressions, the volume fraction of fillers

\[ c = \int_{\Omega} \theta(\mathbf{X}) d\mathbf{X} \]  

(2.19)

has been included as an explicit argument in the effective energy \( \overline{W} \) for later convenience, while \( \mathcal{K}, \mathcal{E} \) denote sufficiently large sets of admissible deformation gradients \( \mathbf{F} \) and curl-free electric fields \( \mathbf{E} \) consistent with the affine boundary conditions (2.15), namely

\[ \mathcal{K} = \{ \mathbf{F} : \exists \mathbf{x} = \chi(\mathbf{X}), \mathbf{F} = \text{Grad} \chi, \det \mathbf{F} > 0, \mathbf{X} \in \Omega, \mathbf{x} = \mathbf{F} \mathbf{X}, \mathbf{X} \in \partial \Omega \}, \]  

(2.20)

and

\[ \mathcal{E} = \{ \mathbf{E} : \exists \Phi = \Phi(\mathbf{X}), \mathbf{E} = -\text{Grad} \Phi, \mathbf{X} \in \Omega, \Phi = -\mathbf{E} \cdot \mathbf{X}, \mathbf{X} \in \partial \Omega \}. \]  

(2.21)
A standard calculation suffices to show that the Euler-Lagrange equations associated with the variational principle (2.18) are exactly the balance of linear momentum and Gauss’s law (2.11). The effective free energy function (2.18) is, by definition, objective in $\mathbf{F}$ and objective and even in $\mathbf{E}$, namely,

$$
W(\mathbf{F}, \mathbf{E}, c) = W(\mathbf{QF}, \mathbf{E}, c) = W(\mathbf{F}, -\mathbf{E}, c)
$$

for all $\mathbf{Q} \in \text{Orth}^+$, and all $\mathbf{F}$ and $\mathbf{E}$, much like its local counterpart $W(\mathbf{X}, \mathbf{F}, \mathbf{E})$. In analogy with the relations (2.7) between the Eulerian and Lagrangian fields, it follows from the definition (2.17)–(2.18) that

$$
\mathbf{T} = \frac{1}{\det \mathbf{F}} \mathbf{SF}^T, \quad \mathbf{d} = \frac{1}{\det \mathbf{F}} \mathbf{FD}, \quad \text{and} \quad \mathbf{p} = \mathbf{d} - \varepsilon_0 \mathbf{F}^{-T} \mathbf{E},
$$

(2.22)

where

$$
\mathbf{T} = \frac{1}{|\Omega_d|} \int_{\Omega_d} \mathbf{T}(\mathbf{x})d\mathbf{x}, \quad \mathbf{d} = \frac{1}{|\Omega_d|} \int_{\Omega_d} \mathbf{d}(\mathbf{x})d\mathbf{x}, \quad \text{and} \quad \mathbf{p} = \frac{1}{|\Omega_d|} \int_{\Omega_d} \mathbf{p}(\mathbf{x})d\mathbf{x},
$$

(2.23)

are the volume averages of the total Cauchy stress $\mathbf{T}$, Eulerian electric displacement $\mathbf{d}$, and polarization $\mathbf{p}$ over the deformed configuration $\Omega_d$. Again, we note that at several passages in this document it will prove useful to treat the macroscopic electric displacement $\mathbf{D}$ as the independent macroscopic electric variable instead of the Lagrangian electric field $\mathbf{E}$. Similar to its local counterpart, details for such a formulation are deferred to Appendix A to avoid loss of continuity.

In general, the solution of the coupled and nonlinear Euler-Lagrange equations (2.11) associated with the variational principal (2.18) is not unique. However, the solution is expected to be unique in a small enough neighborhood of $\mathbf{F} = \mathbf{I}$ and $\mathbf{E} = \mathbf{0}$, with $\mathbf{I}$ denoting the identity in the space of second-order tensors. As the norms of the macroscopic deformation $||\mathbf{F} - \mathbf{I}||$ and macroscopic electric field $||\mathbf{E}||$ are increased, this solution may bifurcate into solutions with different energies. The computation of all such bifurcated solutions is in general an impossibility. Here, following common praxis (see, e.g., Geymonat et al., 1993; Michel et al., 2000), we adopt a semi-inverse approach and restrict the sets $\mathcal{K}$ and $\mathcal{E}$ of admissible deformation gradients and electric fields to include only certain subclasses of fields. This is to exclude potential bifurcated solutions associated with local geometric instabilities that do not impact the macroscopic response of the composite. Formally, instead of considering the variational problem (2.18) per say, this amounts to considering the variational problem

$$
\overline{W}^\sharp(\mathbf{F}, \mathbf{E}, c) = \min_{\mathbf{F} \in \mathcal{K}^\sharp} \max_{\mathbf{E} \in \mathcal{E}^\sharp} \int_{\Omega} W(\mathbf{X}, \mathbf{F}, \mathbf{E})d\mathbf{X},
$$

(2.24)

where $\mathcal{K}^\sharp$ and $\mathcal{E}^\sharp$ are appropriately restricted subsets of $\mathcal{K}$ and $\mathcal{E}$. It follows from this definition that $\overline{W}^\sharp = \overline{W}$ from $\mathbf{F} = \mathbf{I}$ and $\mathbf{E} = \mathbf{0}$ up to the point corresponding to the first bifurcation, after which
\[ W^d \neq \bar{W} \]. To ease the notation, we shall drop the use of the symbol \( \dagger \) in \( \bar{W} \) henceforth, with the understanding that \( \bar{W} \) stands for the solution of the variational problem \( (2.24) \).

### 2.2.1 The classical limit of small deformations and moderate electric fields

The macroscopic constitutive response \( (2.17)–(2.18) \) simplifies significantly in the classical limit of small macroscopic deformations and moderate macroscopic electric fields.\(^4\) (see, e.g., Section 2.25 in Stratton, 1941; Toupin, 1956). In this limit, defining \( \zeta \) as a vanishingly small parameter, the deformation measure \( H = F - I \) is assumed to be \( O(\zeta) \) while the electric field \( E \) is assumed to be \( O(\zeta^{1/2}) \), and the effective free energy function \( (2.18) \) reduces asymptotically to

\[
W(F, E, c) = \frac{1}{2} H_{ij} \hat{L}_{ijkl}(c) H_{kl} - \frac{1}{2} \epsilon_{ij}(c) E_j + H_{ij} \hat{M}_{ijkl}(c) E_k E_l - E_i E_j \hat{\tau}_{ijkl}(c) E_k E_l + O(\zeta^3). \tag{2.25}
\]

Here, \( \hat{L} \) stands for the effective modulus of elasticity, \( \hat{\epsilon} \) denotes the effective permittivity, \( \hat{M} \) is the effective electrostrictive tensor, and \( \hat{\tau} \) represents the effective permittivity of second order. Because of the energy character and overall objectivity of the definition \( (2.18) \), these tensors exhibit the symmetries \( \hat{L}_{ijkl} = \hat{L}_{klij}, \hat{\epsilon}_{ij} = \hat{\epsilon}_{ji}, \hat{M}_{ijkl} = \hat{M}_{jikl}, \hat{\tau}_{ijkl} = \hat{\tau}_{jikl} = \hat{\tau}_{kjil} = \hat{\tau}_{ljki} = \hat{\tau}_{ikjl} = \hat{\tau}_{ilkj} = \hat{\tau}_{ijlk} \). The corresponding relations for the macroscopic stress and electric displacement \( (2.17) \) reduce to

\[
S_{ij} = \frac{\partial W}{\partial F_{ij}}(F, E, c) = \hat{L}_{ijkl}(c) H_{kl} + \hat{M}_{ijkl}(c) E_k E_l + O(\zeta^2) \tag{2.26}
\]

and

\[
D_i = -\frac{\partial W}{\partial E_i}(F, E, c) = \hat{\epsilon}_{ij}(c) E_j + O(\zeta^{3/2}) \tag{2.27}
\]

to leading order. Here, it is important to recognize that the asymptotic constitutive relation \( (2.26) \) for the stress is \( O(\zeta) \) while the asymptotic constitutive relation \( (2.27) \) for the electric displacement is of different order, \( O(\zeta^{1/2}) \). It is also important to recognize that the permittivity of second order \( \hat{\tau} \) does not enter in either expression \( (2.26) \) or \( (2.27) \). That is, in this classical limit of small macroscopic deformations and moderate macroscopic electric fields, the overall elastic dielectric response of dielectric elastomer composites is characterized by three effective tensors: the fourth-order tensor \( \hat{L} \) describing their elasticity, the second-order tensor \( \hat{\epsilon} \) describing their permittivity, and the fourth-order \( \hat{M} \) tensor describing their electrostrictive response.

\(^4\)For conciseness, we may occasionally refer to this very limit in the rest of this document as ‘limit of small deformations’ or ‘small-deformation limit’.
2. The Problem

Remarkably, in spite of the inherent coupling and nonlinearity of the problem, it is possible to write formulae for the effective elastic dielectric tensors \( \tilde{L}, \tilde{\epsilon}, \tilde{M} \) solely in terms of a purely elastic problem and an uncoupled purely dielectric problem (Tian et al., 2012). With help of the notation

\[
L(X) = [1 - \theta(X)]L^{(1)} + \theta(X)L^{(2)},
\]
\[
\epsilon(X) = [1 - \theta(X)]\epsilon^{(1)} + \theta(X)\epsilon^{(2)},
\]
\[
M(X) = [1 - \theta(X)]M^{(1)} + \theta(X)M^{(2)},
\]

(2.28)
(2.29)
(2.30)

for the local elastic modulus \( L \), local permittivity tensor \( \epsilon \), and local electrostrictive tensor \( M \) of the composite, with the elastic moduli \( L^{(r)} \), permittivity tensors \( \epsilon^{(r)} \), and electrostrictive tensors \( M^{(r)} \) of the matrix (\( r = 1 \)) and particles (\( r = 2 \))

\[
L^{(r)} = \frac{\partial^2 W^{(r)}}{\partial F^2} (I, 0), \quad \epsilon^{(r)} = \frac{\partial^2 W^{(r)}}{\partial E^2} (I, 0), \quad M^{(r)} = \frac{1}{2} \frac{\partial^3 W^{(r)}}{\partial F \partial E^2} (I, 0),
\]

(2.31)

these formulae read for a two-phase dielectric elastomer composite as (Tian et al., 2012)

\[
\tilde{L}_{ijkl} = \int_{\Omega} L_{ijrs} \Gamma_{rkl,s} dX,
\]
\[
\tilde{\epsilon}_{ij} = \int_{\Omega} \epsilon_{is} \gamma_{s,j} dX,
\]
\[
\tilde{M}_{ijkl} = \int_{\Omega} \Gamma_{rij,s} M_{rsuv} \gamma_{u,k} \gamma_{v,l} dX,
\]

(2.33)
(2.34)
(2.35)

where the third- and first-order tensor fields \( \Gamma \) and \( \gamma \) are defined implicitly as the solutions to the following linear uncoupled boundary value problems:

\[
[L_{ijrs} \Gamma_{rkl,s}]_{j} = 0, \quad X \in \Omega \quad \text{with} \quad \Gamma_{ikl} = \delta_{ik}\delta_{jl}X_{j}, \quad X \in \partial\Omega
\]

(2.36)

and

\[
[\epsilon_{is}\gamma_{s,j}]_{i} = 0, \quad X \in \Omega \quad \text{with} \quad \gamma_{i} = \delta_{ij}X_{j}, \quad X \in \partial\Omega.
\]

(2.37)

Here and subsequently, the notation \( \cdot \) represents partial differentiation with respect to the material point coordinate \( X_{i} \), \( \delta_{ij} \) denotes the Kronecker delta, and it is recalled that \( \partial\Omega \) stands for the boundary of the domain \( \Omega \).

5This formulation applies more generally to any dielectric elastomer composite characterized by a free-energy function \( W(X, F, E) \). In this context, the local elastic modulus \( L \), local permittivity tensor \( \epsilon \), and local electrostrictive tensor \( M \) of the composite are given by

\[
L(X) = \frac{\partial^2 W}{\partial F^2} (X, I, 0), \quad \epsilon(X) = \frac{\partial^2 W}{\partial E^2} (X, I, 0), \quad M(X) = \frac{1}{2} \frac{\partial^3 W}{\partial F \partial E^2} (X, I, 0),
\]

(2.32)

and are to be used in the formulation (2.33)–(2.37).
A general closed-form solution in the small-deformation limit for a general class of iterative microstructures

Some people create with words, or with music, or with a brush and paints. I like to make something beautiful when I run. I like to make people stop and say, “I’ve never seen anyone run like that before”. It’s more than just a race, it’s style. It’s doing something better than everyone else. It’s being creative.

– Steve Prefontaine, quoted by Don Chapman, sportswriter for the Daily Emerald

This chapter is the first of four addressing the homogenization problem for dielectric elastomer composites in the limit of small deformations and moderate electric fields as introduced in Section 2.2.1. Here, we generate a rigorous analytical solution for the macroscopic electromechanical response of dielectric elastomer composites with a special but fairly general class of random and periodic two-phase particulate microstructures. Namely, closed-form formulas are derived for the three effective electromechanical tensors $\tilde{L}$, $\tilde{\epsilon}$, and $\tilde{M}$ characterizing the macroscopic elastic dielectric response of the composites directly in terms of the corresponding tensors describing the electromechanical response of the underlying matrix and particles, and the one- and two-point correlation functions describing the size, shape and spatial distribution of the latter. This is accomplished by specializing a new iterative homogenization theory in finite electroelastostatics (Lopez-Pamies, 2014) to the case of elastic dielectrics with even coupling between the mechanical and electric fields and, subsequently, carrying out the pertinent asymptotic analysis. Motivated by practical applications,
3. A general closed-form solution in the small-deformation limit

we record the “specialization” of the derived solution to the cases of dielectric elastomers filled with transversely isotropic distributions of aligned spheroidal particles, isotropic distributions of spherical particles, and transversely isotropic distributions of aligned fibers with circular cross section.

3.1 The constitutive theory of Lopez-Pamies (2014) for elastic dielectric composites

By means of a combination of iterative techniques, Lopez-Pamies (2014) has recently generated an exact solution for the variational problem (2.18) for two-phase elastic dielectric composites with a specific, yet fairly general, class of particulate microstructures. In the present notation, his result for the effective free energy function $W = W(F, E, c)$ is given implicitly by the Hamilton-Jacobi (HJ) partial differential equation (pde)

$$c \frac{\partial W}{\partial c} - W - \int_{|\xi|=1} \max_{\alpha} \min_{\beta} \left[ \alpha \cdot \frac{\partial W}{\partial F} \xi + \beta \frac{\partial W}{\partial E} \cdot \xi - W^{(1)}(F + \alpha \otimes \xi, E + \beta \xi) \right] \nu(\xi) d\xi = 0 \quad (3.1)$$

subject to the initial condition

$$W(F, E, 1) = W^{(2)}(F, E), \quad (3.2)$$

where the integration of the pde (3.1) is to be carried out from $c = 1$ to the desired final value of volume fraction of particles $c = c$ and the weighting function $\nu(\xi)$ in (3.1) is given in terms of the two-point correlation function

$$p^{(2)}(Y) = \int_{\Omega} \theta(Y + X) \theta(X) dX \quad (3.3)$$

as follows:

- **Random microstructures.** For the case of random distributions of particles, the function $\nu(\xi)$ is given by

$$\nu(\xi) = -\frac{1}{8\pi^2} \int_{\Omega} p^{(2)}(X) - c^2 \delta''(\xi \cdot X) dX, \quad (3.4)$$

1The two-point correlation function $p^{(22)}(Y)$ represents the probability that the ends of a rod of length and orientation described by the vector $Y$ land within (the same or two different) particles when dropped randomly in $\Omega$ (see, e.g., Chapter 15 in Milton, 2002 and references therein). We recall that the volume fraction $c$ of particles corresponds to the one-point correlation function $p^{(2)} = \int_{\Omega} \theta(X) dX$, that is, the probability that a point lands in a particle when it is dropped randomly in $\Omega$. As opposed to $p^{(2)}$, $p^{(22)}$ contains information about the shape and spatial distribution of the particles in the undeformed configuration.
where \( \delta'' \) denotes the second derivative of the Dirac delta function with respect to its argument.

Direct use of relation (3.4) allows to rewrite the pde (3.1) more explicitly as

\[
\frac{c}{\partial } \frac{\partial W}{\partial c} - \bar{W} + \frac{1}{8\pi^2} \int_{|\xi|=1} \frac{1}{\Omega} \max_{\alpha} \min_{\beta} \left[ \alpha \cdot \frac{\partial W}{\partial \mathbf{F}} \cdot \xi + \beta \frac{\partial W}{\partial \mathbf{E}} \cdot \xi - \delta''(\mathbf{x} \cdot \mathbf{X}) \right] p^{(22)}(\mathbf{X}) (1 - c) c \frac{\partial}{\partial \mathbf{x} \cdot \mathbf{X}} = 0.
\]

(3.5)

• **Periodic microstructures.** For the case of periodic distributions of particles, the function \( \nu(\xi) \) is given by

\[
\nu(\xi) = \sum_{\mathbf{k} \in \mathbb{R}^* \setminus \{0\}} \frac{\tilde{\nu}^{(22)}(\mathbf{k})}{(1 - c) c} \delta \left( \xi - \frac{\mathbf{k}}{\mathbf{|k|}} \right) \quad \text{with} \quad \tilde{\nu}^{(22)}(\mathbf{k}) = \frac{1}{|Q|} \int_{Q} p^{(22)}(\mathbf{X}) e^{-i\mathbf{x} \cdot \mathbf{X}} d\mathbf{X}.
\]

(3.6)

Here, \( \delta(\xi - \mathbf{k}/|\mathbf{k}|) \) denotes the Dirac delta function and \( \tilde{\nu}^{(22)}(\mathbf{k}) \) stands for the Fourier transform of the two-point correlation function \( p^{(22)}(\mathbf{X}) \), while \( Q \) denotes the repeating unit cell chosen to describe the microstructure and

\[
\mathcal{R}^* = \{ \mathbf{k} : \mathbf{k} = n_1 \mathbf{B}_1 + n_2 \mathbf{B}_2 + n_3 \mathbf{B}_3, \quad n_i \in \mathbb{Z} \},
\]

(3.7)

with

\[
\mathbf{B}_1 = 2\pi \frac{\mathbf{A}_2 \wedge \mathbf{A}_3}{\mathbf{A}_1 \cdot (\mathbf{A}_2 \wedge \mathbf{A}_3)}, \quad \mathbf{B}_2 = 2\pi \frac{\mathbf{A}_3 \wedge \mathbf{A}_1}{\mathbf{A}_1 \cdot (\mathbf{A}_2 \wedge \mathbf{A}_3)}, \quad \mathbf{B}_3 = 2\pi \frac{\mathbf{A}_1 \wedge \mathbf{A}_2}{\mathbf{A}_1 \cdot (\mathbf{A}_2 \wedge \mathbf{A}_3)}.
\]

(3.8)

stands for the reciprocal lattice in Fourier space of the periodic lattice in real space

\[
\mathcal{R} = \{ \mathbf{X} : \mathbf{X} = n_1 \mathbf{A}_1 + n_2 \mathbf{A}_2 + n_3 \mathbf{A}_3, \quad n_i \in \mathbb{Z} \}
\]

(3.9)

over which the unit cell \( Q \) is repeated. Upon invoking the identity \( \tilde{\nu}^{(22)}(\mathbf{k}) = |\tilde{\nu}^{(2)}(\mathbf{k})|^2 \) with

\[
\tilde{\nu}^{(2)}(\mathbf{k}) = |Q|^{-1} \int_{Q} \theta^{(2)}(\mathbf{X}) e^{-i\mathbf{x} \cdot \mathbf{X}} d\mathbf{X},
\]

the pde (3.1) adopts the more explicit form

\[
\frac{c}{\partial } \frac{\partial W}{\partial c} - \bar{W} - \sum_{\mathbf{k} \in \mathbb{R}^* \setminus \{0\}} \max_{\alpha} \min_{\beta} \left[ \alpha \cdot \frac{\partial W}{\partial \mathbf{F}} \cdot \xi + \beta \frac{\partial W}{\partial \mathbf{E}} \cdot \xi - \delta''(\mathbf{x} \cdot \mathbf{X}) \right] p^{(22)}(\mathbf{X}) (1 - c) c \frac{\partial}{\partial \mathbf{x} \cdot \mathbf{X}} = 0.
\]

(3.10)

The interested reader is referred to Lopez-Pamies (2014) for the derivation and thorough discussion of the above result. At this stage, nevertheless, it is appropriate to remark that the result (3.1)–(3.2) is exact for a specific class of two-phase particulate microstructures and hence it is realizable. Moreover, it is valid for arbitrary free energy functions \( W^{(1)} \) and \( W^{(2)} \) and arbitrary electroelastic coupling, such as piezoelectric materials (Spinelli and Lopez-Pamies, 2014), as well as with even electroelastic coupling, such as the dielectric elastomers of interest here.

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\(^2\)This theory allows as well to consider elastic dielectric matrix and particles with odd electroelastic coupling, such as piezoelectric materials (Spinelli and Lopez-Pamies, 2014), as well as with even electroelastic coupling, such as the dielectric elastomers of interest here.
one-point $p^{(2)}$ and two-point $p^{(22)}$ correlation functions (3.3). Given this generality, the result (3.1)–(3.2) can be utilized more broadly as a constitutive theory for two-phase elastic dielectrics with *any* particulate microstructure: for a given matrix constitutive behavior $W^{(1)}$, given particle constitutive behavior $W^{(2)}$, and given one- and two-point correlation functions $p^{(2)}$ and $p^{(22)}$, the result (3.1)–(3.2) provides a constitutive model for the macroscopic response of the elastic dielectric composite of interest.

### 3.2 Application to dielectric elastomer composites

By virtue of their synthesis and fabrication process into a network of long polymeric chains coiled randomly without a preferred direction, most elastomers are mechanically and dielectrically isotropic. Typically, they are also such that their polarization is proportional to the applied electric field but independent of the applied deformation, much like liquid polymers (see, e.g., Kofod et al., 2003; Wissler and Mazza, 2007; Di Lillo et al., 2011). For our purposes then, based on these observations together with the fact that the ultimate goal of this chapter is to examine the limit of small deformations and moderate electric fields, it suffices to restrict attention (without loss of generality) to dielectric elastomer composites wherein the matrix material is characterized by a free energy function of the form

$$W^{(1)}(\mathbf{F}, \mathbf{E}) = \frac{\mu}{2} [\mathbf{F} \cdot \mathbf{F} - 3] - \mu (J - 1) + \frac{\lambda + \mu}{2} (J - 1)^2 - \frac{\varepsilon}{2} \mathbf{F}^{-T} \mathbf{E} \cdot \mathbf{F}^{-T} \mathbf{E}. \quad (3.11)$$

Here, $\mathbf{F} \cdot \mathbf{F} = F_{ij} F_{ij}$, $J = \det \mathbf{F}$, and the parameters $\mu$, $\lambda$, $\varepsilon$ stand for the initial Lamé constants and permittivity of the elastomeric material under consideration. On the other hand, the filler particles in dielectric elastomer composites may possibly be anisotropic, both mechanically and dielectrically. That is the case, for instance, for single crystal and textured ceramic particles. No restriction is hence made on the free energy function $W^{(2)}$, other than, again, its even functional dependence on the electric field.

Now, for the matrix free energy function (3.11), the maximizing vector $\alpha$ and minimizing scalar
3. A general closed-form solution in the small-deformation limit

$\beta$ in (3.1) can be determined in closed form. They read as

$$
\alpha = \frac{1}{\mu} \frac{\partial W}{\partial F} \xi - \overline{F} \xi + \frac{J}{\mu} \left[ \frac{(\lambda + 2\mu) - J(\lambda + \mu)}{\mu + J^2(\lambda + \mu)F^{-T} \xi \cdot F^{-T} \xi} \right] F^{-T} \xi + \frac{1}{\mu} \left( \frac{\partial W}{\partial E} \cdot \xi \right) \overline{F}^{-T} \overline{E}^{-T} \xi$$

$$
\left( \frac{\partial W}{\partial E} \cdot \xi \right) \left( \frac{\partial W}{\partial E} \cdot \xi \right) \frac{2 \varepsilon J F^{-T} \xi \cdot F^{-T} \xi}{\mu F^{-T} \xi \cdot F^{-T} \xi} F^{-T} \xi -$$

$$
\left( \frac{\partial W}{\partial E} \cdot \xi \right) \left( \frac{\partial W}{\partial E} \cdot \xi \right) \left( \frac{1}{J} \frac{\partial W}{\partial E} \right) \left( \frac{1}{J} \frac{\partial W}{\partial E} \right) F^{-T} \xi -$$

$$
- \left( \frac{\partial W}{\partial E} \cdot \xi \right) \left( \frac{\partial W}{\partial E} \cdot \xi \right) \left( \frac{1}{J} \frac{\partial W}{\partial E} \right) \left( \frac{1}{J} \frac{\partial W}{\partial E} \right) F^{-T} \xi$$

(3.12)

and

$$\beta = \alpha \cdot F^{-T} \overline{E} - \frac{1 + \alpha \cdot F^{-T} \xi}{F^{-T} \xi \cdot F^{-T} \xi} \left[ \frac{1}{J} \frac{\partial W}{\partial E} \xi + F^{-T} \xi \cdot F^{-T} \xi \right],$$

(3.13)

where $\mathcal{J} \equiv \det \mathbf{F}$ has been introduced to ease notation. Direct use of relations (3.12)–(3.13) in expression (3.1), together with some lengthy but straightforward calculations, allows to write the solution for the effective free energy function in the more explicit form

$$
\overline{W}(F, E, c) = \overline{U}(F, E, c) + \frac{\mu}{2} (F \cdot F - 3) - \mu(J - 1) + \frac{\lambda + \mu}{2} (J - 1)^2 - \frac{\varepsilon}{2} \mathcal{J} F^{-T} \xi \cdot F^{-T} \xi,
$$

(3.14)

where the function $\overline{U}$ is defined implicitly as the solution to the pde

$$
\varepsilon \frac{\partial \overline{U}}{\partial \xi} - \overline{U} - \int_{|\xi| = 1} \frac{1}{2 \mu} \frac{\partial \overline{U}}{\partial F} \xi \cdot \frac{\partial \overline{U}}{\partial F} \xi + \frac{J^2(\lambda + \mu)}{2 \mu} \left( \frac{\partial \overline{U}}{\partial F} \xi \cdot F^{-T} \xi \right)^2 + \frac{1}{\mu} \left( \frac{\partial \overline{U}}{\partial E} \cdot \xi \right) \times
$$

$$
\left[ \frac{\partial \overline{U}}{\partial E} \xi \cdot F^{-T} \xi - \frac{J^2(\lambda + \mu)}{\mu + J^2(\lambda + \mu)F^{-T} \xi \cdot F^{-T} \xi} \left( \frac{\partial \overline{U}}{\partial F} \xi \cdot F^{-T} \xi \right) \right] +
$$

$$
\frac{\left( \frac{\partial \overline{U}}{\partial E} \cdot \xi \right)^3}{8 \varepsilon J^2 F^{-T} \xi \cdot F^{-T} \xi} \left[ \mu + J^2(\lambda + \mu)F^{-T} \xi \cdot F^{-T} \xi \right] -
$$

$$
\left( \frac{\partial \overline{U}}{\partial E} \cdot \xi \right)^2 \left[ \mu + J^2(\lambda + \mu)F^{-T} \xi \cdot F^{-T} \xi \right] + \frac{1}{2 \mu} \left( \frac{\partial \overline{U}}{\partial E} \cdot \xi \right)^2 \overline{F}^{-T} \overline{E}^{-T} \overline{F}^{-T} \overline{E}^{-T} \xi$$

$$
+ \left( \frac{\partial \overline{U}}{\partial E} \cdot \xi \right)^2 \mathcal{J}(\lambda + \mu) \left[ \mu + \varepsilon J \left( \overline{F}^{-T} \overline{E}^{-T} \xi \cdot \overline{F}^{-T} \overline{E}^{-T} \xi \right)^2 \right]
$$

$$
\frac{1}{2 \varepsilon \mu} \left[ \mu + J^2(\lambda + \mu)F^{-T} \xi \cdot F^{-T} \xi \right] \nu(\xi) d\xi = 0.
$$

(3.15)
subjected to the initial condition
\[
U(F, E, 1) = W^{(2)}(F, E) - \frac{\mu}{2} [F \cdot F - 3] + \mu (J - 1) - \frac{\lambda + \mu}{2} (J - 1)^2 + \frac{\varepsilon}{2} J F^T E \cdot F^T E.
\] (3.16)

The computation of the effective free energy function \(W\) amounts thus to solving the initial-value problem (3.15)–(3.16) for \(U\). In view of the polynomial nonlinearity of the pde (3.15), this initial-value problem might admit a closed-form solution, at least for some choices of particle constitutive behaviors and microstructures (see, e.g., Lopez-Pamies et al., 2013a). The analytical solvability and properties of the pde (3.15) in its general form, however, is a substantial task deferred for future work. In the rest of this chapter, we restrict our attention to its asymptotic behavior in the limit of small deformations and moderate electric fields.

### 3.3 Asymptotic solution in the limit of small deformations and moderate electric fields

The result (3.14) with (3.15)–(3.16) for the effective free energy function \(W\) of dielectric elastomer composites is valid for arbitrarily large macroscopic deformation gradients \(F\) and arbitrarily large macroscopic electric fields \(E\). In this section, we examine its asymptotic behavior in the classical limit of small deformations and moderate electric fields, as described in Section 2.2.1.

Before proceeding with the pertinent details, we make use of the isotropic nature of the matrix but keep the behavior of the particles general, so that (2.31) specialize to
\[
L^{(1)} = 2\mu K + (3\lambda + 2\mu) J, \quad \epsilon^{(1)} = \varepsilon I, \quad M^{(1)} = \varepsilon K - \frac{\varepsilon}{2} J, \quad (3.17)
\]
\[
L^{(2)} = \frac{\partial^2 W^{(2)}}{\partial F^2}(I, 0), \quad \epsilon^{(2)} = -\frac{\partial^2 W^{(2)}}{\partial E^2}(I, 0), \quad M^{(2)} = \frac{1}{2} \frac{\partial^3 W^{(2)}}{\partial F \partial E^2}(I, 0), \quad (3.18)
\]
and introduce for later convenience the following quantities:
\[
\Delta L = L^{(2)} - L^{(1)}, \quad \Delta \epsilon = \epsilon^{(2)} - \epsilon^{(1)}, \quad \Delta M = M^{(2)} - M^{(1)}, \quad (3.19)
\]
where the tensors \(K, J\) are given in component form by
\[
K_{ijkl} = \frac{1}{2} \left[ \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl} \right], \quad J_{ijkl} = \frac{1}{3} \delta_{ij} \delta_{kl}, \quad (3.20)
\]
and again \(\delta_{ij}\) denotes the Kronecker delta. For later reference, we remark that \(K\) and \(J\) are orthogonal projection tensors with the properties \(K J = J K = 0, K K = K, J J = J, K + J = I,\)
where $\mathcal{I}$ stands for the identity in the space of fourth-order tensors with major and minor symmetries, $\mathcal{I}_{ijkl} = 1/2 (\delta_{ik}\delta_{jl} + \delta_{ij}\delta_{jk})$. We recall from Section 2.2.1 that the tensors $L^{(r)}$, $e^{(r)}$, $M^{(r)}$ in (3.17)–(3.18) correspond physically to the modulus of elasticity, the permittivity, and the electrostrictive free energy function (3.14), where the effective tensors $W^{(r)}$ (3.18) correspond physically to the modulus of elasticity, the permittivity, and the electrostrictive system of differential equations associated with powers $I^r$.

A general closed-form solution in the small-deformation limit 25 initial condition (3.16) about the system of coupled nonlinear ordinary differential equations (odes)

$$c \frac{d\tilde{L}_{ijkl}}{dc} - \Delta \tilde{L}_{ijkl} - \Delta \tilde{L}_{ijpq}P_{pqmn}^{L} \Delta \tilde{L}_{mnkl} = 0,$$

$$c \frac{d\tilde{e}_{ij}}{dc} - \Delta \tilde{e}_{ij} - \Delta \tilde{e}_{ij}P_{pq}^{e} \Delta \tilde{e}_{qj} = 0,$$

$$c \frac{d\tilde{M}_{ijkl}}{dc} - \Delta \tilde{M}_{ijkl} - \Delta \tilde{L}_{ijpq}P_{pqmn}^{L} \Delta \tilde{M}_{mnkl} - \Delta \tilde{M}_{ijkl}P_{pq}^{e} \Delta \tilde{e}_{qj} - \Delta \tilde{M}_{ijkl}P_{pq}^{e} \Delta \tilde{e}_{qj} + Q_{ijkl}^{M} = 0,$$

subject to the initial conditions

$$\tilde{L}(1) = L^{(2)}, \quad \tilde{e}(1) = e^{(2)}, \quad \tilde{M}(1) = M^{(2)}.$$

In the above expressions, we have made use of the notation $\Delta \tilde{L} = \tilde{L} - L^{(1)}$, $\Delta \tilde{e} = \tilde{e} - e^{(1)}$, $\Delta \tilde{M} = \tilde{M} - M^{(1)}$, and $P^{L}$, $P^{e}$, $Q^{M}$ are microstructural tensors given in component form by

$$P_{ijkl}^{L} = \frac{1}{\mu} \delta_{ik} \langle \xi_j \xi_l \rangle |_{(ij),(kl)} - \frac{\mu + \lambda}{\mu(2\mu + \lambda)} \langle \xi_i \xi_j \xi_k \xi_l \rangle,$$

$$P_{ij}^{e} = \frac{1}{\varepsilon} \langle \xi_i \xi_j \rangle,$$

$$Q_{ijkl}^{M} = \frac{1}{2\varepsilon(2\mu + \lambda)} \Delta \tilde{L}_{ijpq} \Delta \tilde{e}_{mk} \Delta \tilde{e}_{nl} \langle \xi_p \xi_q \xi_m \xi_n \rangle + \frac{1}{\varepsilon} \Delta \tilde{e}_{pk} \Delta \tilde{e}_{ql} \langle \xi_i \xi_j \xi_p \xi_q \rangle - \frac{1}{2\varepsilon} \Delta \tilde{e}_{pk} \Delta \tilde{e}_{ql} \delta_{ij} \langle \xi_p \xi_q \rangle - \frac{\lambda + \mu}{\mu(2\mu + \lambda)} \Delta \tilde{L}_{ijpq} \Delta \tilde{e}_{mk} \langle \xi_p \xi_q \xi_m \xi_l \rangle |_{(kl)} + \frac{1}{\mu} \Delta \tilde{L}_{ijpq} \Delta \tilde{e}_{kl} \langle \xi_p \xi_q \rangle |_{(kl)},$$

where the bracketed subscripts imply symmetrization and the symbol

$$\langle \cdot \rangle \doteq \int_{|\xi|=1} \nu(\xi) d\xi$$

(3.28)
has been introduced for further notational simplicity; recall from Section 3.1 that the weighting function \( \nu(\xi) \) is given directly in terms of the two-point correlation function \( p^{(22)} \) by expression (3.4) for random microstructures and by (3.6) for periodic ones.

From a computational point of view, it is useful to recognize that the odes (3.21) and (3.22), with initial conditions (3.24)\(_1\) and (3.24)\(_2\), for the effective modulus of elasticity \( \tilde{L} \) and effective permittivity \( \tilde{\epsilon} \) are nonlinear Riccati equations uncoupled from each other and from the ode (3.23) for the effective electrostrictive tensor \( \tilde{M} \). In spite of their quadratic nonlinearity, they can be solved in closed form. Their solutions read simply as

\[
\tilde{L}_{ijkl} = \mu(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \lambda\delta_{ij}\delta_{kl} + c \left\{ [(1 - c)P^L + \Delta L^{-1}]^{-1} \right\}_{ijkl} \tag{3.29}
\]

and

\[
\tilde{\epsilon}_{ij} = \varepsilon\delta_{ij} + c \left\{ [(1 - c)P^e + \Delta \epsilon^{-1}]^{-1} \right\}_{ij}. \tag{3.30}
\]

Having determined the results (3.29) and (3.30), the linear ode (3.23), which does depend on \( \tilde{L} \) and \( \tilde{\epsilon} \), can also be solved in closed form to render

\[
\tilde{M}_{ijkl} = \frac{\varepsilon}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \delta_{ij}\delta_{kl}) + \frac{1}{c^2} \Delta \tilde{L}_{ijpq} \Delta L^{-1}_{mnsp} \Delta M_{pqrs} \Delta \epsilon_{vr}^{-1} \Delta \epsilon_{uk} \Delta \epsilon_{sv}^{-1} \Delta \epsilon_{vl} - \\
\frac{\varepsilon^2 - 1}{4c^2\varepsilon(2\mu + \lambda)} \Delta \tilde{L}_{ijpq} \Delta \epsilon_{kr} \Delta \epsilon_{ts} \langle \xi_p \xi_q \xi_r \xi_s \rangle + \frac{1 - c}{2c\varepsilon} \Delta \tilde{\epsilon}_{kr} \Delta \tilde{\epsilon}_{ts} \left[ \langle \xi_p \xi_q \xi_r \xi_s \rangle - \frac{1}{2} \delta_{ij} \langle \xi_r \xi_s \rangle \right] - \\
\frac{(\lambda + \mu)/(1 - c)}{2c^2\mu(2\mu + \lambda)} \Delta \tilde{L}_{ijpq} \left[ c \delta_{mk} + \Delta \epsilon_{mr}^{-1} \Delta \epsilon_{kr} \right] \Delta \epsilon_{ts} \langle \xi_p \xi_q \xi_m \xi_s \rangle \left|_{(kl)} \right. + \\
\frac{1 - c}{2c^2\varepsilon} \Delta \tilde{L}_{ijpq} \Delta L_{pqmn}^{-1} \Delta \tilde{\epsilon}_{kr} \Delta \tilde{\epsilon}_{ts} \left[ \langle \xi_m \xi_n \xi_r \xi_s \rangle - \frac{1}{2} \delta_{mn} \langle \xi_r \xi_s \rangle \right]. \tag{3.31}
\]

The exact closed-form solutions (3.29), (3.30), (3.31) constitute the main result of this chapter. They characterize the overall electromechanical response of dielectric elastomer composites with a large class of random and periodic particulate microstructures in the limit of small deformations and moderate electric fields. The following theoretical and practical remarks are in order:

\( i. \) Elastic dielectric behaviors of the matrix and particles. The solutions (3.29), (3.30), (3.31) are valid for any choice of Lamé constants \( \mu, \lambda \) and permittivity \( \varepsilon \) describing the isotropic elastic dielectric behavior of the elastomeric matrix, as well as for any choice of modulus of elasticity \( \mathbf{L}^{(2)} \), permittivity \( \varepsilon^{(2)} \), and electrostrictive tensor \( \mathbf{M}^{(2)} \) describing the (possibly anisotropic) elastic dielectric behavior of the underlying particles.
For the practically relevant case when the particles are isotropic ideal elastic dielectrics with
\[ L^{(2)} = 2\mu_p K + (3\lambda_p + 2\mu_p) J, \quad \epsilon^{(2)} = \varepsilon_p I, \quad M^{(2)} = \varepsilon_p K - \frac{\varepsilon_p}{\mu} J, \tag{3.32} \]
where \( \mu_p, \lambda_p, \varepsilon_p \) denote the Lamé constants and permittivity of the particles, the solutions (3.29), (3.30), (3.31) specialize to
\[ \tilde{L}_{ijkl} = \mu (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \lambda \delta_{ij}\delta_{kl} + c \left\{ \left[ (1-c) P^I + \frac{1}{2\Delta\mu} K + \frac{1}{3\Delta\lambda + 2\Delta\mu} J \right]^{-1} \right\}_{ijkl}, \tag{3.33} \]
\[ \bar{\epsilon}_{ij} = \varepsilon \delta_{ij} + c \left\{ \left[ (1-c) P^I + \frac{1}{\Delta\varepsilon} I \right]^{-1} \right\}_{ij}, \tag{3.34} \]
\[ \bar{M}_{ijkl} = \frac{\varepsilon}{2} (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \delta_{ij}\delta_{kl}) + \frac{1}{2\mu^2 \Delta\mu \Delta\varepsilon} \Delta \tilde{L}_{ijpq} \Delta \tilde{\epsilon}_{kp} \Delta \tilde{\epsilon}_{iq} + \frac{1-c}{2\varepsilon} \Delta \tilde{\epsilon}_{kr} \Delta \tilde{\epsilon}_{is} \langle \xi_i \xi_j \xi_k \xi_s \rangle - \frac{\Delta \lambda + \Delta \mu}{2 \varepsilon} \Delta \tilde{L}_{ijm} \Delta \tilde{\epsilon}_{km} \Delta \tilde{\epsilon}_{rl} - \frac{1-c}{4 \varepsilon} \Delta \tilde{\epsilon}_{kr} \Delta \tilde{\epsilon}_{is} \delta_{ij} \langle \xi_i \xi_s \rangle - \frac{1-c}{4 \varepsilon} \Delta \tilde{L}_{ijpq} \Delta \tilde{\epsilon}_{pk} \Delta \tilde{\epsilon}_{is} \langle \xi_p \xi_q \xi_i \xi_s \rangle \mid_{(kl)} - \frac{1}{2 \mu^2 \Delta \mu} \left\{ \frac{\varepsilon^2 - 1}{2 \varepsilon (2\mu + \lambda)} - \frac{1-c}{\mu (2\mu + \lambda)} + (\lambda + \mu) \right\} \Delta \tilde{L}_{ijpq} \Delta \tilde{\epsilon}_{kr} \Delta \tilde{\epsilon}_{is} \langle \xi_p \xi_q \xi_k \xi_s \rangle - \frac{1-c}{2 \varepsilon \mu (2\mu + \lambda)} \Delta \tilde{L}_{ijpq} \Delta \tilde{\epsilon}_{is} \langle \xi_p \xi_q \xi_k \xi_s \rangle \mid_{(kl)} + \frac{1-c}{2 \varepsilon \mu} \Delta \tilde{L}_{ijpq} \langle \xi_p \xi_q \xi_k \xi_s \rangle \mid_{(kl)}, \tag{3.35} \]
where use has been made of the notation \( \Delta \mu = \mu_p - \mu, \Delta \lambda = \lambda_p - \lambda, \Delta \varepsilon = \varepsilon_p - \varepsilon \), and the fact that \( K \) and \( J \) are orthogonal projection tensors.

ii. Geometry and spatial distribution of the particles. The solutions (3.29), (3.30), (3.31) are also valid for any choice of the one- and two-point correlation functions \( p^{(2)} = c \) and \( p^{(22)} \) describing the microstructure (see e.g., equation (3.3)). In practice, both of these quantities are generally measurable and often times readily known from the outset. In particular, the information on \( p^{(22)} \) enters the solutions (3.29), (3.30), (3.31) through the microstructural tensors \( \langle \xi \otimes \xi \rangle \) and \( \langle \xi \otimes \xi \otimes \xi \otimes \xi \rangle \). For demonstration purposes and later use, we spell out next the specialization of these tensors to two basic cases: (i) a random distribution of aligned ellipsoidal particles and (ii) a periodic distribution of ellipsoidal particles. Figure 3.1 depicts schematics of these two microstructures with the various quantities of interest indicated.
3. A general closed-form solution in the small-deformation limit

Figure 3.1: (a) Schematic of a random distribution of aligned ellipsoidal particles. (b) Rectangular prismatic unit cell, with principal axes $\mathbf{u}_1, \mathbf{u}_2, \mathbf{u}_3$ and sides $b_1, b_2, b_3$, describing a periodic cuboidal distribution of ellipsoidal particles.

For microstructures comprised of aligned ellipsoidal particles whose centers are distributed randomly with the so-called “ellipsoidal” symmetry introduced by Willis (1977), the weighting function (3.4) can be determined explicitly allowing to write

$$
\langle \xi_i \xi_j \xi_k \xi_l \rangle = \frac{\det \mathbf{Z}}{4\pi} \int_{|\xi|=1} \frac{\xi_i \xi_j \xi_k \xi_l}{|Z\xi|^3} \, d\xi,
$$

$$
\langle \xi_i \xi_j \rangle = \frac{\det \mathbf{Z}}{4\pi} \int_{|\xi|=1} \frac{\xi_i \xi_j}{|Z\xi|^3} \, d\xi.
$$

Expressions (3.36) contain three limiting cases worth remarking. Setting $\mathbf{Z} = \mathbf{I}$ corresponds to an isotropic distribution of spherical particles. Taking the limit of one of the axes of the ellipsoidal particles to be infinitely long corresponds to a distribution of aligned cylindrical fibers with elliptical cross section. Taking the limit of two of the axes of the ellipsoidal particles to be infinitely long corresponds, in turn, to a random distribution of aligned layers (see, e.g., Section 2.13 in Lopez-Pamies, 2006 for technical details on these limits).

For microstructures comprised of a periodic distribution of ellipsoidal particles where the repeating unit cell is a rectangular prism of sides $b_1, b_2, b_3$ containing a single particle located at its center, it is not difficult to compute the Fourier transform (3.6) of its two-point correlation
In these expressions, \( Z \) describes, as in the foregoing, the shape and alignment of the particles, \( c = 4\pi \det Z/3b_1b_2b_3 \), and

\[
\xi = \frac{2\pi}{b_1} p_1 \mathbf{u}_1 + \frac{2\pi}{b_2} p_2 \mathbf{u}_2 + \frac{2\pi}{b_3} p_3 \mathbf{u}_3
\]

in terms of the summation integers \( p_1, p_2, p_3 \), where the mutually orthogonal unit vectors \( \mathbf{u}_1, \mathbf{u}_2, \mathbf{u}_3 \) stand for the principal axes of the unit cell, as depicted in Fig. 3.1(b). Setting \( Z = I \) in (3.37) corresponds to a periodic distribution of spherical particles. The limiting cases of a periodic distribution of aligned cylindrical fibers with elliptical cross section and a periodic distribution of aligned layers are also contained in expressions (3.37).

iii. Connection with the Hashin-Shtrikman variational principles in elastostatics and electrostatics.

By construction, the underlying microstructure associated with the solutions (3.29), (3.30), (3.31) corresponds to a distribution of disconnected particles that interact in such a manner that their deformation gradient and electric field — irrespectively of the value of the volume fraction of particles \( c \) — are uniform and the same in each particle (see Appendix B in Lopez-Pamies, 2014).

An interesting implication of such a special type of intra-particle fields is that the solutions (3.29) and (3.30) for the effective modulus of elasticity \( \tilde{L} \) and effective permittivity \( \tilde{\epsilon} \) agree identically with the variational approximations obtained from the Hashin-Shtrikman variational principles in elastostatics (Hashin and Shtrikman, 1962a) and electrostatics (Hashin and Shtrikman, 1962b) when choosing the reference medium to coincide with the matrix material and the trial polarization field to be constant per phase (so that the fields within the particles are also constant). A corollary of this agreement is that the solutions (3.29) and (3.30) coincide identically with one of the Hashin-Shtrikman bounds when the elastic and dielectric properties of the matrix and particles are well ordered: the result (3.29) agrees with the upper (lower) bound in elastostatics when \( L^{(2)} < L^{(1)} \) (\( L^{(2)} > L^{(1)} \)) in the sense of quadratic forms,
while the result (3.30) agrees with the upper (lower) bound in electrostatics when \( \varepsilon^{(2)} < \varepsilon^{(1)} \) \((\varepsilon^{(2)} > \varepsilon^{(1)})\) also in the sense of quadratic forms. In view of these connections, it would be interesting to explore in future studies whether the solution (3.31) for the effective electrostrictive tensor \( \tilde{M} \) possesses similar extremal properties.

iv. Connection with the classical results for dilute suspensions of ellipsoidal particles in elastostatics and electrostatics. A further implication of the uniformity of the intra-particle fields is that the solutions (3.29) and (3.30) agree with the classical results for the effective modulus of elasticity \( \tilde{L} \) (Eshelby, 1957) and effective permittivity \( \tilde{\varepsilon} \) (see, e.g., Bergman, 1978 and references therein) of a dilute suspension of aligned ellipsoidal particles. By contrast, through a comparison with an exact solution available for spherical particles, we show in the next section that expression (3.31) does not coincide in general with the effective electrostrictive tensor \( \tilde{M} \) of a dilute suspension of aligned ellipsoidal particles.

**Transversely isotropic dielectric elastomer composites.** We focus now on dielectric elastomer composites wherein the fillers are distributed with transverse isotropy in the undeformed configuration \( \Omega \). Due to their microstructure, this type of composites exhibits an overall electromechanical behavior that is transversely isotropic. We shall denote their initial axis of symmetry by the unit vector \( N \); as an illustrative example, Fig. 3.2 shows a schematic of a transversely isotropic dielectric elastomer composite with a distribution of axisymmetric fillers aligned in the direction of the axis of symmetry \( N \). For this class of anisotropic dielectric elastomers, the effective free energy

![Figure 3.2: Schematic of the microstructure of a transversely isotropic dielectric elastomer composite with initial axis of symmetry \( N \).](image-url)
3. A general closed-form solution in the small-deformation limit

function (2.18) satisfies the material symmetry requirement \( \mathcal{W}(\mathbf{F}, \mathbf{K}, \mathbf{E}, c) = \mathcal{W}(\mathbf{F}, \mathbf{E}, c) \) for all \( \mathbf{F}, \mathbf{E} \), and all \( \mathbf{K} \in \text{Symm} \) with \( \text{Symm} = \{ \mathbf{Q}, \mathbf{Q} = \mathbf{R}_N(\theta) \} \), where \( \mathbf{R}_N(\theta) \) denotes a rotation of angle \( \theta \) about the vector \( \mathbf{N} \). By means of standard calculations, it is not difficult to show that this requirement implies that the effective tensors \( \mathbf{\bar{L}}, \mathbf{\bar{e}}, \mathbf{\bar{M}} \) in the macroscopic constitutive relations (2.26)–(2.27) are transversely isotropic tensors, as expected. With help of the Walpole notation (Walpole, 1981), they can then be expediently written as

\[
\mathbf{\bar{L}} = \bar{c}_L \mathbf{E}^{(1)} + \bar{d}_L \mathbf{E}^{(2)} + \bar{\sigma}_L \mathbf{E}^{(3)} + \bar{f}_L \mathbf{E}^{(4)} + \bar{g}_L (\mathbf{E}^{(5)} + \mathbf{E}^{(6)}),
\]

\[
\mathbf{\bar{e}} = \bar{\varepsilon}_t (\mathbf{I} - \mathbf{N} \otimes \mathbf{N}) + \bar{\varepsilon}_l \mathbf{N} \otimes \mathbf{N},
\]

\[
\mathbf{\bar{M}} = \bar{c}_M \mathbf{E}^{(1)} + \bar{d}_M \mathbf{E}^{(2)} + \bar{\sigma}_M \mathbf{E}^{(3)} + \bar{f}_M \mathbf{E}^{(4)} + \bar{g}_M \mathbf{E}^{(5)} + \bar{h}_M \mathbf{E}^{(6)}
\]  

(3.39)

with

\[
\mathbf{E}^{(1)}_{ijkl} = \frac{1}{2}(\delta_{ij} - N_i N_j)(\delta_{kl} - N_k N_l),
\]

\[
\mathbf{E}^{(2)}_{ijkl} = N_i N_j N_k N_l,
\]

\[
\mathbf{E}^{(3)}_{ijkl} = \mathbf{E}^{(1)}_{ikjl} = \mathbf{E}^{(1)}_{jikl},
\]

\[
\mathbf{E}^{(4)}_{ijkl} = \frac{1}{2}(\mathbf{E}^{(5)}_{ikjl} + \mathbf{E}^{(5)}_{jikl} + \mathbf{E}^{(5)}_{iljk} + \mathbf{E}^{(5)}_{ljik} + \mathbf{E}^{(5)}_{jkl}),
\]

\[
\mathbf{E}^{(5)}_{ijkl} = N_i N_j (\delta_{kl} - N_k N_l),
\]

\[
\mathbf{E}^{(6)}_{ijkl} = \mathbf{E}^{(5)}_{klij},
\]

(3.40)

where it is recalled that \( \delta_{ij} \) denotes the Kronecker delta. From these expressions it is trivial to deduce that, in the limit of small deformations and moderate electric fields, the overall electromechanical response of transversely isotropic dielectric elastomers is characterized by thirteen (five elastic\(^3\), two dielectric, and six electrostrictive) effective constants.

Next, we work results for the thirteen effective constants \( \bar{c}_L, \bar{d}_L, \bar{\sigma}_L, \bar{f}_L, \bar{g}_L, \bar{\varepsilon}_t, \bar{\varepsilon}_l, \bar{c}_M, \bar{d}_M, \bar{\sigma}_M, \bar{f}_M, \bar{g}_M, \bar{h}_M \) in (3.39) directly in terms of the electromechanical properties of the matrix and fillers, as characterized by the free energy functions \( W^{(1)} \) and \( W^{(2)} \), and the microstructure, as

\(^3\)In the literature, there are several sets of five constants that are utilized to characterize the linear elastic response of transversely isotropic materials. Perhaps the more standard set is that comprised of the transverse (or in-plane) bulk modulus \( \tilde{\kappa}_t \) and shear modulus \( \tilde{\mu}_t \), and the longitudinal shear modulus \( \tilde{\mu}_l \), Young’s modulus \( \tilde{E}_l \), and Poisson’s ratio \( \tilde{\nu}_l \). These are given in terms of the five elastic constants \( \bar{c}_L, \bar{d}_L, \bar{\sigma}_L, \bar{f}_L, \bar{g}_L \) that appear in the Walpole notation (3.39)\(_1\) by the following relations:

\[
\begin{align*}
\bar{\kappa}_t &= \frac{\bar{c}_L}{2}, & \tilde{\kappa}_t &= \frac{\bar{d}_L}{2}, & \tilde{\mu}_t &= \frac{\bar{f}_L}{2}, & \tilde{E}_l &= \bar{d}_L - 2\frac{\bar{d}_L^2}{\bar{c}_L}, & \tilde{\nu}_l &= \frac{\bar{g}_L}{\bar{c}_L}.
\end{align*}
\]  

(3.41)
characterized by the indicator function $\theta$, for the case when the fillers are spheroidal particles that are aligned and whose centers are distributed with transversely isotropic symmetry.

The solution (3.35) is valid for any choice of the one- and two-point correlation functions $p^{(2)} = c$ and $p^{(22)}$ describing the microstructure. The latter enters expressions (3.35) through the microstructural tensors $\langle \xi_i \xi_j \xi_k \xi_l \rangle$ and $\langle \xi_i \xi_j \rangle$. For later use, it is expedient to recognize that for transversely isotropic microstructures these tensors admit the spectral form

$$\langle \xi_i \xi_j \xi_k \xi_l \rangle = \alpha_1 \left( E^{(1)}_{ijkl} + \frac{1}{2} E^{(3)}_{ijkl} \right) + \alpha_2 E^{(2)}_{ijkl} + \alpha_3 \left( E^{(4)}_{ijkl} + \frac{1}{2} E^{(5)}_{ijkl} + \frac{1}{2} E^{(6)}_{ijkl} \right),$$

$$\langle \xi_i \xi_j \rangle = \beta_1 (\delta_{ij} - N_i N_j) + \beta_2 N_i N_j,$$

(3.42)

Here,

$$\alpha_1 = \frac{1}{2} \langle E^{(1)}_{mmij} \xi_i \xi_j \xi_k \xi_l E^{(1)}_{klnm} \rangle, \quad \alpha_2 = \langle E^{(2)}_{mmij} \xi_i \xi_j \xi_k \xi_l E^{(2)}_{klnm} \rangle, \quad \alpha_3 = \frac{1}{2} \langle E^{(4)}_{mmij} \xi_i \xi_j \xi_k \xi_l E^{(4)}_{klnm} \rangle,$$

$$\beta_1 = \frac{1}{2} (1 - \xi_i N_i \xi_j N_j), \quad \beta_2 = (\xi_i N_i \xi_j N_j),$$

(3.43)

where it is recalled that the tensors $E^{(1)}$ through $E^{(6)}$ are given in terms of the axis of symmetry $N$ by relations (3.40), and that the triangular brackets stand for the weighted average (3.28) with the weight $\nu(\xi)$ being given by expression (3.4) in terms of the two-point correlation function $p^{(22)}$.

For transversely isotropic microstructures, in view of the relations (3.42)-(3.43), the solution (3.35) for the effective electromechanical tensors $\vec{L}$, $\vec{e}$, $\vec{M}$ reduces indeed to the form (3.39) with the thirteen effective electromechanical constants given by

$$\vec{c}_L = 2\lambda + 2\mu + \frac{2(\Delta \lambda + \Delta \mu)c}{\Delta \mu(3\lambda + 2\mu)Y} - \frac{2c(1-c)[\alpha_2(\lambda + \mu) - \beta_2(\lambda + 2\mu)]}{\mu(\lambda + 2\mu)Y},$$

$$\vec{d}_L = \lambda + 2\mu + \frac{(\Delta \lambda + 2\Delta \mu)c}{\Delta \mu(3\lambda + 2\mu)Y} - \frac{2c(1-c)[\alpha_1(\lambda + \mu) - \beta_1(\lambda + 2\mu)]}{\mu(\lambda + 2\mu)Y},$$

$$\vec{e}_L = 2\mu + \frac{2\mu\Delta \mu(\lambda + 2\mu)c}{(\lambda + 2\mu)[\mu + 2(1-c)\beta_1 \Delta \mu] - (1-c)\alpha_1 \Delta \mu(\lambda + \mu)},$$

$$\vec{f}_L = 2\mu + \frac{2\mu\Delta \mu(\lambda + 2\mu)c}{(\lambda + 2\mu)[\mu + (1-c)\Delta \mu(\beta_1 + \beta_2)] - 2(1-c)\alpha_3 \Delta \mu(\lambda + \mu)},$$

$$\vec{g}_L = \lambda + \frac{c\Delta \lambda}{\Delta \mu(3\lambda + 2\mu)Y} + \frac{c(1-c)(\lambda + \mu)\alpha_3}{\mu(\lambda + 2\mu)Y},$$

$$\vec{\epsilon}_t = \epsilon + \frac{c\epsilon \Delta \epsilon}{\epsilon + (1-c)\beta_1 \Delta \epsilon}, \quad \vec{\epsilon}_l = \epsilon + \frac{c\epsilon \Delta \epsilon}{\epsilon + (1-c)\beta_2 \Delta \epsilon},$$

(3.44)
\[\tilde{c}_M = \left(1 - c\right)\Delta \tilde{e}_i^2 (\alpha_1 - \beta_1) + \frac{\Delta \tilde{e}_i^2 \left(\tilde{c}_L - 2\lambda - 2\mu\right)\Delta \lambda - 2(\tilde{g}_L - \lambda)(\Delta \lambda + \Delta \mu)}{2c \varepsilon} + \frac{\Delta \tilde{e}_i^2 \left(\tilde{c}_L - 2\lambda - 2\mu\right)\Delta \lambda - 2(\tilde{g}_L - \lambda)(\Delta \lambda + \Delta \mu)}{2c^2 \Delta \mu \Delta \varepsilon (3\Delta \lambda + 2\Delta \mu)}
\]

\[\tilde{d}_M = \frac{\varepsilon}{2} + \frac{(1 - c)\Delta \tilde{e}_i^2 (2\alpha_2 - \beta_2)}{4c \varepsilon} + \frac{\Delta \tilde{e}_i^2 \left(\tilde{d}_L - \lambda - 2\mu\right)(2\Delta \lambda + \Delta \mu) - 2(\tilde{g}_L - \lambda)(\Delta \lambda + \Delta \mu)}{2c^2 \Delta \mu \Delta \varepsilon (3\Delta \lambda + 2\Delta \mu)} + \frac{\alpha_3(\tilde{g}_L - \lambda)}{4c^2 \varepsilon \Delta \mu} \left(1 + \left(1 + c\right)\Delta \mu \frac{\Delta \lambda + c\Delta \varepsilon}{\mu \Delta \varepsilon \Delta \tilde{e}_i (\Delta \lambda + \Delta \mu)}\right) - \frac{\beta_1(1 - c)\Delta \tilde{e}_i^2 (\Delta \lambda + \Delta \mu)}{2c^2 \varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)} \left[\tilde{g}_L - \lambda + (\tilde{c}_L - 2\lambda - 2\mu) \left(1 - \frac{\varepsilon \Delta \mu (\Delta \tilde{e}_i + c\Delta \varepsilon) (3\Delta \lambda + 2\Delta \mu)}{\mu \Delta \varepsilon \Delta \tilde{e}_i (\Delta \lambda + \Delta \mu)}\right)\right],\]

\[\tilde{e}_M = \varepsilon + \frac{\alpha_1(1 - c)\Delta \tilde{e}_i^2 (\tilde{d}_L - 2\mu)}{4c \varepsilon} + \frac{\Delta \tilde{e}_i^2 \left(\tilde{d}_L - \lambda - 2\mu\right)(2\Delta \lambda + \Delta \mu) - 2(\tilde{g}_L - \lambda)(\Delta \lambda + \Delta \mu)}{2c^2 \Delta \mu \Delta \varepsilon (3\Delta \lambda + 2\Delta \mu)} + \frac{\alpha_3(\tilde{g}_L - \lambda)}{4c^2 \varepsilon \Delta \mu} \left(1 + \left(1 + c\right)\Delta \mu \frac{\Delta \lambda + c\Delta \varepsilon}{\mu \Delta \varepsilon \Delta \tilde{e}_i (\Delta \lambda + \Delta \mu)}\right) - \frac{\beta_1(1 - c)\Delta \tilde{e}_i^2 (\Delta \lambda + \Delta \mu)}{2c^2 \varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)} \left[\tilde{g}_L - \lambda + (\tilde{c}_L - 2\lambda - 2\mu) \left(1 - \frac{\varepsilon \Delta \mu (\Delta \tilde{e}_i + c\Delta \varepsilon) (3\Delta \lambda + 2\Delta \mu)}{\mu \Delta \varepsilon \Delta \tilde{e}_i (\Delta \lambda + \Delta \mu)}\right)\right],\]

\[\tilde{f}_M = \varepsilon + \frac{\alpha_3(1 - c)\Delta \tilde{e}_i^2 (\tilde{f}_L - 2\mu)}{2c \varepsilon} + \frac{\Delta \tilde{e}_i^2 \left(\tilde{f}_L - \lambda - 2\mu\right)(2\Delta \lambda + \Delta \mu) - \Delta \lambda (\tilde{g}_L - \lambda)}{2c^2 \Delta \mu \Delta \varepsilon + 2\Delta \lambda + \Delta \mu} + \frac{\beta_2(1 - c)\Delta \tilde{e}_i^2 (\Delta \lambda + \Delta \mu)}{2c^2 \varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)} \left[\tilde{f}_L - \lambda + (\tilde{d}_L - \lambda - 2\mu) \left(1 - \frac{\varepsilon \Delta \mu (\Delta \tilde{e}_i + c\Delta \varepsilon) (3\Delta \lambda + 2\Delta \mu)}{\mu \Delta \varepsilon \Delta \tilde{e}_i (\Delta \lambda + \Delta \mu)}\right)\right],\]

\[\tilde{g}_M = \varepsilon + \frac{(1 - c)\Delta \tilde{e}_i^2 (\alpha_3 - \beta_1)}{4c \varepsilon} - \frac{\Delta \tilde{e}_i^2 \left(\tilde{g}_L - \lambda - 2\mu\right)(\Delta \lambda + \Delta \mu) - \Delta \lambda (\tilde{g}_L - \lambda)}{2c \varepsilon} + \frac{\alpha_3(\tilde{d}_L - \lambda - 2\mu) + 2\alpha_1(\tilde{g}_L - \lambda)}{4c^2 \varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)} \left[\frac{\lambda + 2\mu + (1 + c)\Delta \mu}{2c^2 \Delta \mu} - \frac{(\lambda + \mu)(\Delta \tilde{e}_i + c\Delta \varepsilon)}{\mu \Delta \varepsilon \Delta \tilde{e}_i}\right] - \frac{\beta_1(1 - c)\Delta \tilde{e}_i^2 (\Delta \lambda + \Delta \mu)}{4c^2 \varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)} \left[\tilde{g}_L - \lambda - 2\mu - 2(\tilde{g}_L - \lambda) \left(1 - \frac{\varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)(\Delta \tilde{e}_i + c\Delta \varepsilon)}{\mu \Delta \varepsilon \Delta \tilde{e}_i (\Delta \lambda + \Delta \mu)}\right)\right],\]

\[\tilde{h}_M = \varepsilon + \frac{(1 - c)\Delta \tilde{e}_i^2 (\alpha_3 - \beta_1)}{4c \varepsilon} - \frac{\Delta \tilde{e}_i^2 \left(\tilde{g}_L - \lambda - 2\mu\right)(\Delta \lambda + \Delta \mu) - (\tilde{g}_L - \lambda)(2\Delta \lambda + \Delta \mu)}{2c \varepsilon} + \frac{\alpha_3(\tilde{g}_L - \lambda - 2\mu) + 2\alpha_2(\tilde{g}_L - \lambda)}{4c^2 \varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)} \left[\frac{\lambda + 2\mu + (1 + c)\Delta \mu}{2c^2 \Delta \mu} - \frac{(\lambda + \mu)(\Delta \tilde{e}_i + c\Delta \varepsilon)}{\mu \Delta \varepsilon \Delta \tilde{e}_i}\right] - \frac{\beta_2(1 - c)\Delta \tilde{e}_i^2 (\Delta \lambda + \Delta \mu)}{4c^2 \varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)} \left[\tilde{g}_L - \lambda - 2\mu - (\tilde{g}_L - \lambda) \left(1 - \frac{2\varepsilon \Delta \mu (3\Delta \lambda + 2\Delta \mu)(\Delta \tilde{e}_i + c\Delta \varepsilon)}{\mu \Delta \varepsilon \Delta \tilde{e}_i (\Delta \lambda + \Delta \mu)}\right)\right],\]

(3.44 cont.)
where the notation $\Delta \tilde{\varepsilon}_l = \tilde{\varepsilon}_l - \varepsilon$, $\Delta \tilde{\varepsilon}_t = \tilde{\varepsilon}_t - \varepsilon$,

$$\Upsilon = \left( \frac{\Delta \lambda + \Delta \mu}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} - \frac{(1 - c)[\alpha_2(\lambda + \mu) - \beta_2(\lambda + 2\mu)]}{\mu(\lambda + 2\mu)} \right) \left( \frac{\Delta \lambda + 2\Delta \mu}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} \right) - \frac{2(1 - c)[\alpha_1(\lambda + \mu) - \beta_1(\lambda + 2\mu)]}{\mu(\lambda + 2\mu)} \right)^2 \right) \right) \right)$$

has been introduced for simplicity.

While specific for transversely isotropic dielectric elastomer composites, expressions (3.44) are still admittedly general as they apply to arbitrary types of transversely isotropic microstructures. Again, the dependence on the microstructure enters via the volume fraction of the fillers $c$ and the five parameters $\alpha_1$, $\alpha_2$, $\alpha_3$, $\beta_1$, $\beta_2$ defined in terms of the two-point correlation function (3.3) by relations (3.43).

In the sequel, we specialize the result (3.44) to dielectric elastomer composites wherein the fillers are aligned spheroidal particles whose centers are distributed with the so-called “spheroidal” symmetry introduced by Willis (Willis, 1977). The two-point correlation function for such a type of microstructures, as characterized by the weight function (3.4), can be determined in closed form. This allows to determine simple formulas for the parameters $\alpha_1$, $\alpha_2$, $\alpha_3$, $\beta_1$, $\beta_2$, and ultimately to write down fully explicit expressions for all thirteen effective constants $\tilde{c}_L$, $\tilde{d}_L$, $\tilde{\varepsilon}_L$, $\tilde{g}_L$, $\tilde{\varepsilon}_t$, $\tilde{\varepsilon}_l$, $\tilde{c}_M$, $\tilde{d}_M$, $\tilde{e}_M$, $\tilde{f}_M$, $\tilde{g}_M$, $\tilde{h}_M$.

### 3.4 Application to dielectric elastomers filled with aligned spheroidal particles

In this section, we work out the “specialization” of the effective electromechanical constants (3.44) to the case of dielectric elastomer composites wherein the underlying fillers are spheroidal in shape and are all aligned in the same direction\(^4\). In order to favor analytical tractability, we consider that the centers of the particles are distributed with “spheroidal” symmetry and that the “aspect ratio” of this spheroidal distribution is the same as the aspect ratio of the particles\(^5\) (Willis, 1977). For this

\(^4\)We emphasize that the microstructure for which the effective free-energy function defined by the Hamilton-Jacobi equation (3.1) is exact comprises particles of infinitely polydisperse size, abstract shape, and spatial distribution. In the sequel, by specialization to aligned spheroidal particles, we mean that the one- and two-point correlation functions in (3.1) are chosen to agree identically with a microstructure wherein the particles are indeed of spheroidal shape and are distributed transverse isotropically.

\(^5\)Approximations (based on classical linear estimates) for the overall electromechanical response of dielectric elastomer composites with this type of microstructures had been proposed earlier by Li and Rao (Li and Rao, 2004) and by Siboni and Ponte Castañeda (Siboni and Ponte Castañeda, 2013), the latter being specific for particles that are mechanically rigid.
class of microstructures, the weight function (3.4) reduces rather simply to (Lopez-Pamies, 2014; Willis, 1977)

\[
\nu(\xi) = \frac{\det Z}{4\pi|Z\xi|^3} \quad \text{with} \quad Z = I + (\omega - 1)N \otimes N.
\]  

(3.46)

Here, the unit vector \(N\), again, denotes the initial orientation of the particles and the non-negative parameter \(\omega\) has been introduced to denote their aspect ratio; it is defined so that the particles are prolate (oblate) for \(\omega > 1\) (\(\omega < 1\)). The parameter \(\omega\) also serves to characterize the spatial distribution of the centers of the particles. Figure 3.3 shows a schematic representation of this class of microstructures, including two of the limits that it contains: an isotropic distribution of spherical particles (corresponding to \(\omega = 1\)) and a transversely isotropic distribution of aligned cylindrical fibers with circular cross section (corresponding to \(\omega = \infty\)).

Figure 3.3: Schematic of the microstructure of a dielectric elastomer filled with aligned spheroidal particles of aspect ratio \(\omega\) distributed with “spheroidal” symmetry of the same aspect ratio \(\omega\). The lower half of the figure shows the limiting cases of (a) an isotropic distribution of spherical particles (corresponding to \(\omega = 1\)) and (b) a transversely isotropic distribution of aligned cylindrical fibers with circular cross section (corresponding to \(\omega = \infty\)).

Now, given the weight function (3.46), it is straightforward to determine the corresponding
parameters (3.43) needed in the evaluation of the effective constants (3.44). They read as follows:

\[
\alpha_1 = \frac{\omega^2(2\omega^2 + 1) - \rho(\omega)(4\omega^2 - 1)}{4(\omega^2 - 1)^2}, \quad \alpha_2 = \frac{2 + \omega^2 - 3\rho(\omega)}{2(\omega^2 - 1)^2}, \quad \alpha_3 = \frac{\rho(\omega)(2\omega^2 + 1) - 3\omega^2}{2(\omega^2 - 1)^2},
\]

\[
\beta_1 = \frac{\omega^2 - \rho(\omega)}{2(\omega^2 - 1)}, \quad \beta_2 = \frac{\rho(\omega) - 1}{\omega^2 - 1},
\]

where

\[
\rho(\omega) = \frac{\omega \sin^{-1}\left(\frac{\sqrt{\omega^2 - 1}}{\sqrt{1 - \omega^2}}\right)}{\sqrt{\omega^2 - 1}} = \omega \sin^{-1}\left(\frac{\sqrt{1 - \omega^2}}{\sqrt{1 - \omega^2}}\right). \quad (3.47)
\]

To ease notation, the explicit dependence of the function \(\rho\) — written here in two different equivalent forms for convenience — on \(\omega\) is dropped in the sequel.

Substitution of relations (3.47) in expressions (3.44) renders the main result of this chapter, namely, the thirteen effective constants \(\tilde{c}_L, \tilde{d}_L, \tilde{e}_L, \tilde{f}_L, \tilde{g}_L, \tilde{\varepsilon}_t, \tilde{\varepsilon}_t, \tilde{c}_M, \tilde{d}_M, \tilde{e}_M, \tilde{f}_M, \tilde{g}_M\) in the three effective tensors (3.39) that characterize the overall electromechanical response of dielectric elastomers filled with a transversely isotropic distribution of aligned spheroidal particles:

\[
\tilde{c}_L = 2\lambda + 2\mu + \frac{2(\Delta \lambda + \Delta \mu) c}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} \tilde{\gamma} + \frac{c(1-c)[2\mu - (3\lambda + 5\mu)\omega^2 + (2\omega^2(\lambda + 2\mu) + \lambda - \mu)\rho]}{\mu(\lambda + 2\mu)(\omega^2 - 1)^2},
\]

\[
\tilde{d}_L = \lambda + 2\mu + \frac{(\Delta \lambda + 2\Delta \mu) c}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} \tilde{\gamma} + \frac{c(1-c)[2\mu\omega^4 - (3\lambda + 5\mu)\omega^2 + (\lambda + 3\mu + 2\lambda \omega^2)\rho]}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2},
\]

\[
\tilde{e}_L = 2\mu + \frac{8\mu c \Delta \mu(\lambda + 2\mu)(\omega^2 - 1)^2}{4\mu(\lambda + 2\mu)(\omega^2 - 1)^2 + (1-c)\Delta \mu [\lambda (3\rho + 2\omega^4 - 5\omega^2) + \mu (7\rho - (4\rho + 9)\omega^2 + 6\omega^4)]},
\]

\[
\tilde{f}_L = 2\mu + \frac{8\mu c \Delta \mu(\lambda + 2\mu)(\omega^2 - 1)^2}{4\mu(\lambda + 2\mu)(\omega^2 - 1)^2 + (1-c)\Delta \mu [\lambda (3\rho + 2\omega^4 - 5\omega^2) + \mu (7\rho - (4\rho + 9)\omega^2 + 6\omega^4)]},
\]

\[
\tilde{g}_L = \lambda + \frac{c \Delta \lambda}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} \tilde{\gamma} + \frac{c(1-c)(\lambda + \mu)[\rho + (2\rho - 3)\omega^2]}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2},
\]

\[
\tilde{\varepsilon}_t = \varepsilon + \frac{2c\varepsilon\Delta \varepsilon (\omega^2 - 1)}{2(\omega^2 - 1)\varepsilon + (1-c)(\omega^2 - \rho)\Delta \varepsilon}, \quad \tilde{\varepsilon}_t = \varepsilon + \frac{c\varepsilon\Delta \varepsilon (\omega^2 - 1)}{(\omega^2 - 1)\varepsilon + (1-c)(\rho - 1)\Delta \varepsilon};
\]

\[
\tilde{c}_M = \frac{\Delta \varepsilon^2 ((\tilde{c}_L - 2\lambda - 2\mu)\Delta \lambda - 2(\tilde{g}_L - \lambda)(\Delta \lambda + \Delta \mu))}{2^2 \Delta \varepsilon (3\Delta \lambda + 2\Delta \mu)} - \frac{(1-c)\Delta \varepsilon^2 (2\rho - 3)\omega^2 + \rho}{4c\varepsilon(\omega^2 - 1)} - \frac{(\Delta \lambda + \Delta \mu)(\omega^2 - \rho)}{c\Delta \mu(3\Delta \lambda + 2\Delta \mu)} \left[ \tilde{g}_L - \lambda + (\tilde{c}_L - 2\lambda - 2\mu) \left( 1 - \varepsilon \Delta \varepsilon \frac{(\lambda + \mu)(\Delta \varepsilon + \Delta \varepsilon t)}{\mu(\lambda + \mu)(\Delta \varepsilon + \Delta \varepsilon t)} \right) \right] - \frac{\omega(2\omega^2 + 1) - \rho(4\omega^2 - 1)}{4c\varepsilon (\omega^2 - 1)} \left[ 1 + (1+c)\Delta \mu \left( 1 - \frac{2\varepsilon(\lambda + \mu)(\Delta \varepsilon + \Delta \varepsilon t)}{(1+c)\mu(\lambda + \mu)(\Delta \varepsilon + \Delta \varepsilon t)} \right) \right] \times \left[ \tilde{c}_L - 2\lambda - 2\mu + \frac{2(\tilde{g}_L - \lambda)[\rho(2\omega^2 + 1) - 3\omega^2]}{\omega^2(2\omega^2 + 1) - \rho(4\omega^2 - 1)} \right].
\]

(3.49)
3. A general closed-form solution in the small-deformation limit

\[
\hat{M} = \varepsilon + \frac{(1 - c)\Delta \tilde{e}_1^2}{2} \left[ \omega^2 (2\omega^2 + 1) + \rho (4\omega^2 - 1) \right] + \frac{\Delta \tilde{e}_1^2}{2\varepsilon \Delta \tilde{e}_1} \left\{ \begin{array}{l}
2\omega^2 + 1 - \rho (\omega^2 + 2) \\
2\omega^2 - 3\rho + 2 \\
\omega^2 + 3\rho - 2
\end{array} \right\} + \frac{(1 - c)\Delta \tilde{e}_1^2}{4\varepsilon (\omega^2 - 1)} \times \\
\left\{ \begin{array}{l}
2\omega^2 + 1 - \rho (\omega^2 + 2) \\
2\omega^2 - 3\rho + 2 \\
\omega^2 + 3\rho - 2
\end{array} \right\} \times \\
\left\{ \begin{array}{l}
\frac{2\omega^2 + 1 - \rho (\omega^2 + 2)}{\omega^2 - 1} \\
\frac{2\omega^2 - 3\rho + 2}{2\varepsilon \Delta \mu (\omega^2 - 1)} \\
\omega^2 + 3\rho - 2
\end{array} \right\} \times \\
\left\{ \begin{array}{l}
(1 + c)\Delta \mu \\
\lambda + 2\mu \\
(1 + c)\mu \Delta \varepsilon \Delta \tilde{e}_1
\end{array} \right\} \\
\left\{ \begin{array}{l}
(1 - \varepsilon)(\lambda + \mu)(\Delta \tilde{e}_1 + c\Delta \varepsilon) \\
\varepsilon \Delta \varepsilon \Delta \tilde{e}_1(\Delta \lambda + \Delta \mu)
\end{array} \right\}
\right)
\right) + \\
\left( \begin{array}{l}
\frac{\Delta \tilde{e}_1^2}{2\varepsilon \Delta \mu \Delta \varepsilon} \left\{ \begin{array}{l}
2\omega^2 + 1 - \rho (\omega^2 + 2) \\
2\omega^2 - 3\rho + 2 \\
\omega^2 + 3\rho - 2
\end{array} \right\} \\
2\varepsilon \Delta \mu (\omega^2 - 1)
\end{array} \right\} \times \\
\left\{ \begin{array}{l}
(1 - c)\Delta \mu \\
\lambda + 2\mu \\
(1 + c)\mu \Delta \varepsilon \Delta \tilde{e}_1
\end{array} \right\} \\
\left\{ \begin{array}{l}
(1 - \varepsilon)(\lambda + \mu)(\Delta \tilde{e}_1 + c\Delta \varepsilon) \\
\varepsilon \Delta \varepsilon \Delta \tilde{e}_1(\Delta \lambda + \Delta \mu)
\end{array} \right\}
\right)
\right)
\right)
\right)
\right)
\right)
where now
\[
Y = \left( \frac{\Delta \lambda + \Delta \mu}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} + \frac{(1-c)\left[ \rho \left( 2\omega^2(\lambda + 2\mu) + \lambda - \mu \right) - (3\lambda + 5\mu)\omega^2 + 2\mu \right]}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2} \right) \\
\times \\
\left( \frac{\Delta \lambda + 2\Delta \mu}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} + \frac{(1-c)[2\mu\omega^2 - (3\lambda + 5\mu)\omega^2 + \rho \left( 2\lambda\omega^2 + \lambda + 3\mu \right)]}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2} \right) \\
- \\
\left( \frac{\Delta \lambda}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} + \frac{(1-c)(\lambda + \mu)\left[ \rho \left( 2\omega^2 + 1 \right) - 3\omega^2 \right]}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2} \right)^2,
\] (3.50)
and it is recalled that \( \Delta \lambda = \lambda_p - \lambda, \Delta \mu = \mu_p - \mu, \Delta \varepsilon = \varepsilon_p - \varepsilon, \Delta \bar{\varepsilon} = \bar{\varepsilon}_p - \bar{\varepsilon}, \) and \( \Delta \bar{\varepsilon} = \bar{\varepsilon}_p - \bar{\varepsilon} \).

There are several limiting cases contained in the result (3.49) that are worth recording explicitly.

- **Rigid particles and incompressible elastomers.** In the limit of rigid particles when \( \mu_p = \lambda_p = \infty \) and incompressible elastomers when \( \lambda = \infty \), the effective electromechanical constants (3.49) reduce to

\[
\bar{c}_L = \bar{d}_L = \infty, \quad \bar{e}_L = 2\mu + \frac{8c\mu(\omega^2 - 1)^2}{(1-c)(3\rho + 2\omega^2 - 5\omega^2)},
\]
\[
\bar{f}_L = 2\mu + \frac{4\epsilon(\omega^2 - 1)^2}{(1-c)(\omega^2 + 1)(2 + \omega^2 - 3\rho)}, \quad \bar{g}_L = \infty;
\]
\[
\bar{\varepsilon}_L = \varepsilon + \frac{2\epsilon\Delta \varepsilon(\omega^2 - 1)}{2(\omega^2 - 1)\epsilon + (1-c)(\omega^2 - \rho)\Delta \varepsilon}; \quad \bar{\varepsilon}_L = \varepsilon + \frac{c\epsilon\Delta \varepsilon(\omega^2 - 1)}{(\omega^2 - 1)\epsilon + (1-c)(\rho - 1)\Delta \varepsilon};
\]
\[
\bar{c}_M = \frac{\Delta \bar{\varepsilon}_t^2}{4\epsilon \Delta \bar{\varepsilon}_t} \left[ 1 + c + \frac{2\varepsilon(\Delta \bar{\varepsilon}_t + c\Delta \varepsilon)}{\Delta \bar{\varepsilon}_t}\right] + \frac{(1+3c)\rho - \omega^2(4\rho + c - 5) - 2(1+c)}{2(\omega^2 - 1)^2},
\]
\[
\bar{d}_M = \bar{c}_L = \infty,
\]
\[
\bar{e}_M = \epsilon + \frac{\Delta \bar{\varepsilon}_t^2}{2c} \left[ \frac{\Delta \bar{\varepsilon}_t + c\Delta \varepsilon}{\Delta \bar{\varepsilon}_t} + \frac{(2\rho - 3\omega^2 - 3 - c)\rho + \omega^2 + 2}{2\epsilon(\omega^2 - 1)^2} \right],
\]
\[
\bar{f}_M = \epsilon + \frac{1}{2} \left( \frac{\Delta \bar{\varepsilon}_t + \omega^2 \Delta \bar{\varepsilon}_t}{\omega^2 + 1} + \frac{\Delta \bar{\varepsilon}_t + c\Delta \varepsilon}{c\Delta \varepsilon} \right) + \frac{(1-c)\Delta \bar{\varepsilon}_t \Delta \bar{\varepsilon}_t \left[ \rho(2\omega^2 + 1) - 3\omega^2 \right]}{4\epsilon \omega^2 (\omega^2 - 1)^2},
\]
\[
\bar{g}_M = \epsilon + \frac{\Delta \bar{\varepsilon}_t^2}{2} \left[ \frac{\omega^2(2\rho - 4\epsilon \rho + c - 3) + (1 + c)\rho + 2c}{8\epsilon \omega^2 (\omega^2 - 1)^2} \right],
\]
\[
\bar{h}_M = -\frac{\epsilon}{2} + \frac{\Delta \bar{\varepsilon}_t^2}{8\epsilon \omega^2 (\omega^2 - 1)^2} \left[ \omega^2(2\rho - 4\epsilon \rho + c - 3) + (1 + 5c)\rho + 4c \right].
\] (3.51)

These results are relevant for standard dielectric elastomers, which are typically nearly incompressible, filled with ceramic or metallic particles.

- **Liquid-like particles and incompressible elastomers.** The limiting values \( \mu_p = 0, \lambda_p = \infty, \) and \( \lambda = \infty \) correspond to particles that are liquid-like (incompressible with vanishingly small
shear resistance) and elastomers that are incompressible. Granted these values, the effective electromechanical constants (3.49) reduce to

\[ \tilde{c}_L = \tilde{d}_L = \infty, \quad \tilde{e}_L = 2\mu - \frac{8c\mu (\omega^2 - 1)^2}{2(1+c)\omega^2 - (3+5c)\omega^2 - 3(1-c)\rho + 4}, \]

\[ \tilde{f}_L = 2\mu - \frac{4c\mu (\omega^2 - 1)^2}{3(1-c)\rho (\omega^2 + 1) + (1+c)\omega^2 - (7-3c)\omega^2 + 2c}, \quad \tilde{g}_L = \infty; \]

\[ \tilde{e}_t = \epsilon + \frac{2c\varepsilon \Delta \varepsilon (\omega^2 - 1)}{2(\omega^2 - 1)\varepsilon + (1-c)(\omega^2 - \rho) \Delta \varepsilon}, \quad \tilde{f}_t = \epsilon + \frac{c\epsilon \Delta \varepsilon (\omega^2 - 1)}{(\omega^2 - 1)\varepsilon + (1-c)(\rho - 1) \Delta \varepsilon}; \]

\[ \tilde{c}_M = \frac{\Delta \tilde{e}_t^2 (\rho - 1)}{2c\varepsilon} \left\{ 1 + \frac{1+c}{2(\rho-1)} - \frac{2c\varepsilon \Delta \varepsilon (\omega^2 - 1)}{(\omega^2 - 1)\varepsilon + (1-c)(\omega^2 - \rho) \Delta \varepsilon} \left[ \frac{1}{2} \frac{\Delta \tilde{e}_t}{\Delta \varepsilon_t} \left[ \frac{1}{2} \frac{c\Delta \varepsilon (\omega^2 - 1) [3 + c(2\rho - 5)]}{8[(1-c)\Delta \varepsilon (\rho - 1) + (\omega^2 - 1)\varepsilon]^2} \left( 1 + \frac{3(1-c) [2(3c-2) (2\omega^2 + 1) \rho^2 + (13 - 9c (4\omega^2 + 1) + 20\omega^2) \rho + 3(9c - 5)\omega^2 - 6]}{[3 + c(2\rho - 5)] [2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1-c)(2\omega^2 + 1)]} \right) \right] \right] \right\} \]

\[ \tilde{d}_M = \frac{\varepsilon + \frac{\Delta \tilde{e}_t^2}{c\Delta \varepsilon} \left\{ 1 + \frac{c\Delta \varepsilon^2 (\omega^2 - 1)^2 [3 + c(2\rho - 5)]}{8[(1-c)\Delta \varepsilon (\rho - 1) + (\omega^2 - 1)\varepsilon]^2} \left( 1 + \frac{3(1-c) [2(3c-2) (2\omega^2 + 1) \rho^2 + (13 - 9c (4\omega^2 + 1) + 20\omega^2) \rho + 3(9c - 5)\omega^2 - 6]}{[3 + c(2\rho - 5)] [2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1-c)(2\omega^2 + 1)]} \right) \right\} \times \]

\[ \left[ 1 - \frac{1+c}{2(1+c)} \frac{3+c}{8(\omega^2 - 1)^2} \right] + \frac{(1-c)(1-c)(\omega^2 - \rho) + 2(3+c)\omega^4 - (5c + 11)\omega^2]{16c(\omega^2 - 1)^2 [4 - 3(1-c)\rho + 2(1+c)\omega^4 - (3+5c)\omega^2]}, \]

\[ \tilde{e}_M = \varepsilon + \frac{4\Delta \tilde{e}_t^2 (\omega^2 - 1)^2}{c\Delta \varepsilon \left[ 4(\omega^2 - 1)^2 - (1-c)(3\rho + 2\omega^4 - 5\omega^2) \right]} \times \]

\[ \left[ 1 - \frac{1+c}{2(1+c)} \frac{3+c}{8(\omega^2 - 1)^2} \right] + \frac{2(1-c)(\omega^2 + \rho - 2\omega^4) \left[ 8 - 3(1-c)\rho + 2(3+c)\omega^4 - (5c + 11)\omega^2 \right]}{16c(\omega^2 - 1)^2 [4 - 3(1-c)\rho + 2(1+c)\omega^4 - (3+5c)\omega^2]}, \]

\[ \tilde{f}_M = \varepsilon - \frac{(1-c)(\Delta \tilde{e}_t)^2}{2(1+c)} + \frac{3+c}{2c(1+c)\Delta \varepsilon} \left\{ \frac{1-c}{2c(1+c)\Delta \varepsilon} \left[ \frac{c(\Delta \tilde{e}_t)^2 (\omega^2 - 1)^2 (7-c-4\rho)}{2(1+c) [(\omega^2 - 1)\varepsilon + (1-c)\Delta \varepsilon (\rho - 1)] [2c(\omega^2 - 1) + (1-c)\Delta \varepsilon (\omega^2 - \rho)]} \left( 1 + \frac{9 (1-c)^2 \rho^2 - 3\rho \left[ 7(\omega^2 - (2 + c) (\omega^2 + 3) + 5] + [27 - c(14 + c)](\omega^2 - 2c(5 + c) \right]}{(7-c-4\rho) \left( 1 + c(\omega^2 + 2c - \omega^2 [4 - 3(1-c)\rho - 3(1-c)(\omega^2 - \rho)] \right) \right) \right) \right\} (3.52) \]
A general closed-form solution in the small-deformation limit

Porous dielectric elastomers.

\[ \tilde{g}_M = -\frac{\varepsilon}{2} + \frac{\Delta \tilde{\varepsilon}_t}{4} - \frac{c\varepsilon \Delta \varepsilon^2}{4} \left( \omega^2 - 1 \right) \left[ 9 + c(8\rho - 11) - 6\rho \right] \left\{ 1 + \frac{3(1-c)}{9 + c(8\rho - 11) - 6\rho} \left[ (2(3c - 2)(2\omega^2 + 1)\rho^2 + \rho \left( 13 - 9c \left( 4\omega^2 + 1 \right) + 20\omega^2 \right) + 3(9c - 5)\omega^2 - 6 \right] \right\} - \frac{\Delta \tilde{\varepsilon}_t^2}{4c\Delta \varepsilon} \left\{ 1 + \frac{\Delta \varepsilon}{2\varepsilon} \left[ 4c^2\rho + c - 3\rho + 2 \right] - c(\rho + 1) + (1 - 3c)\omega^4 \right\} \right\}, \]

\[ \tilde{h}_M = -\frac{\varepsilon}{2} + \frac{\Delta \tilde{\varepsilon}_t^2(\rho - 1)}{2c\varepsilon} \left\{ \frac{\omega^2(2\rho - 1) - 1}{4(\rho - 1)(\omega^2 - 1)^2} + \frac{\Delta \varepsilon^2 \Delta \tilde{\varepsilon}_t^2}{\Delta \varepsilon \Delta \tilde{\varepsilon}_t^2} \left[ 2c(\rho - 1) - 2c + 2\omega^2 + 1 \right] - 2\varepsilon (\omega^2 - 1) \left( \Delta \tilde{\varepsilon}_t - c\Delta \varepsilon \right) \right\} \frac{\Delta \tilde{\varepsilon}_t^2 \left[ 2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1 - c)(2\omega^2 + 1) \right]}{\Delta \varepsilon \Delta \tilde{\varepsilon}_t^2 \left[ 2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1 - c)(2\omega^2 + 1) \right]}, \]

\[ (3.52 \text{ cont.}) \]

The results (3.52) are relevant for standard dielectric elastomers filled with common fluids, such as water, or eutectic alloys, such as Galinstan.

- Porous dielectric elastomers. Another limit of practical relevance contained in the result (3.49) is that of porous dielectric elastomers. This corresponds to setting \( \mu_p = \lambda_p = 0 \) and \( \varepsilon_p = \varepsilon_0 \), with \( \varepsilon_0 = 8.85 \times 10^{-12} \text{ F/m} \) denoting the permittivity of vacuum. Assuming that the underlying elastomer is incompressible, so that \( \lambda = \infty \), the effective electromechanical constants (3.49) in this context reduce to

\[ \tilde{c}_L = \frac{2(1 - c)\mu}{c} \left[ 1 + \frac{(2\rho - 1) \left( 3\rho - \omega^2 - 2 \right) - 6\rho(\rho - 1)^2}{3(1-c)(2\omega^2 + 1) \rho - (5-9c)\omega^2 - 2\omega^4 - 2} \right], \]

\[ \tilde{d}_L = \frac{(1-c)\mu}{c} \left[ \frac{6(1-c)\rho^2 + 4\rho \left( 1 + 3c \right) \omega^2 - 1 - 2(1 + 3c)\omega^4 - 4\omega^2}{3(1-c)(2\omega^2 + 1) \rho - (5-9c)\omega^2 - 2\omega^4 - 2} \right], \]

\[ \tilde{e}_L = 2\mu + \frac{8c\mu \left( \omega^2 - 1 \right)^2}{3(1-c)\rho - 2(1 + c)\omega^4 + 3 + 5c)\omega^2 - 4}, \]

\[ \tilde{f}_L = 2\mu - \frac{4c\mu \left( \omega^2 - 1 \right)^2}{3(1-c)(\omega^2 + 1) \rho + (1 + c)\omega^4 - (7 - 3c)\omega^2 + 2c}, \]

\[ \tilde{g}_L = \frac{2(1-c)\mu}{c} \left[ \frac{3(1-c)\rho^2 + \rho \left( 2 - 3c \right) \omega^2 - 2 - 2(1 - 3c)\omega^2 - 2\omega^4}{3(1-c)(2\omega^2 + 1) \rho - (5-9c)\omega^2 - 2\omega^4 - 2} \right]; \]

\[ \tilde{\varepsilon}_t = \varepsilon + \frac{2c\varepsilon \Delta \varepsilon \left( \omega^2 - 1 \right)}{2(\omega^2 - 1)\varepsilon + (1 - c)(\omega^2 - \rho) \Delta \varepsilon}, \quad \tilde{\varepsilon}_l = \varepsilon + \frac{c\varepsilon \Delta \varepsilon \left( \omega^2 - 1 \right)}{(\omega^2 - 1)\varepsilon + (1 - c)(\rho - 1) \Delta \varepsilon}; \]

\[ (3.53) \]
\[
\bar{c}_M = -(1 - c)\Delta\bar{\ep}_t^2 \left\{ \Delta\bar{\ep}_t + c\Delta\ep + \frac{4 + 6(1 + c)\omega^4 + [2 + 9c]\rho - \omega^2[2(\rho + 5) + 15c]}{24c^2\Delta\ep (\omega^2 - 1)^2} \times \frac{\varepsilon\Delta\ep^2(2\rho - 5)(\omega^2 - 1)}{6\Delta\bar{\ep}_t^2[(1 - c)(\omega^2 - \rho)\Delta\ep + 2(\omega^2 - 1)\varepsilon^2]} \times \left[ 1 + \frac{9\left[2 + (2\rho - 3)\varepsilon\rho - (3 - c)\rho + \omega^2\right]}{(2\rho - 5)[2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1 - c)(2\omega^2 + 1)]} \right] \right\}
\]

\[
\bar{d}_M = \varepsilon - \frac{(1 - 2c)\Delta\bar{\ep}_t^2}{2c^2\Delta\ep} \left\{ 1 + \frac{3c(1 - c)\Delta\ep(\rho - 1)}{2c(1 - 2c)(\omega^2 - 1)^2} \left[ 1 - \frac{(\rho^2 - 1)[1 + 9c - (1 + 6c)\rho]}{9c(\rho - 1)} \right] \right\} - \frac{9(3\rho - \omega^2 - 2) - 27c^2(2\rho - 3)\left[\rho(2\omega^2 + 1) - 3\omega^2\right]}{[3c(2\rho - 5) + 4\rho - 1][2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1 - c)(2\omega^2 + 1)]} \times \frac{18c\left[\rho(3\rho - 8)(2\omega^2 + 1) + 12\omega^2 + 3\right]}{[3c(2\rho - 5) + 4\rho - 1][2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1 - c)(2\omega^2 + 1)]} \times \frac{(1 - c)\Delta\ep^2(\omega^2 - 1)[3c(2\rho - 5) + 4\rho - 1]}{24[(1 - c)\Delta\ep(\rho - 1) + (\omega^2 - 1)\varepsilon^2]},
\]

\[
\bar{e}_M = \varepsilon + \frac{3c(1 - c)\Delta\ep}{c\Delta\ep} \left( \frac{4(\omega^2 - 1)^2 - (1 - c)(3\rho + 2\omega^4 - 5\omega^2)}{8(\omega^2 - 1)^2} \right) + \frac{(1 - c)\Delta\ep^2((1 - 4\rho)\omega^2 + \rho + 2\omega^4)}{16c(\omega^2 - 1)^2[4 - 3(1 - c)\rho + 2(1 + c)\omega^4 - (3 + 5c)\omega^2]} \times \left[ 1 - \left(1 + \frac{c\Delta\ep}{\Delta\ep} \right) \right] + \frac{(1 - c)\Delta\ep(\omega^2 - 1)[(1 - c)\Delta\ep(\rho - 1)]}{(7 - c - 4\rho)} \times 2(1 + c)\left[(\omega^2 - 1)\varepsilon + (1 - c)\Delta\ep(\rho - 1)\right] \left[2c(\omega^2 - 1) + (1 - c)\Delta\ep(\omega^2 - \rho)\right] \times \left[ 1 + \frac{9(1 - c)^2\rho^2 - 3\rho^2[7\omega^2 - c(2 + c)(\omega^2 + 3) + 5 + 27 - c(14 + c)]\omega^2 - 2c(5 + c)}{(7 - c - 4\rho)(1 + c)\omega^4 + 2c - \omega^2[4 - 3(1 - c)\rho] - 3(1 - c)(\omega^2 - \rho)} \right],
\]

\[
\bar{f}_M = \varepsilon - \frac{(1 - c)\Delta\bar{\ep}_t^2}{2(1 + c)} + \frac{(3 + c)\Delta\bar{\ep}_t\Delta\bar{\ep}_t}{2c(1 + c)\Delta\ep} + \frac{(1 - c)\Delta\bar{\ep}_t\Delta\bar{\ep}_t}{4c(\omega^2 - 1)^2} \times \frac{c(1 - c)\varepsilon\Delta\ep(\omega^2 - 1)(7 - c - 4\rho)}{2(1 + c)\left[(\omega^2 - 1)\varepsilon + (1 - c)\Delta\ep(\rho - 1)\right] \left[2c(\omega^2 - 1) + (1 - c)\Delta\ep(\omega^2 - \rho)\right] \times \left[ 1 + \frac{9(1 - c)^2\rho^2 - 3\rho^2[7\omega^2 - c(2 + c)(\omega^2 + 3) + 5 + 27 - c(14 + c)]\omega^2 - 2c(5 + c)}{(7 - c - 4\rho)(1 + c)\omega^4 + 2c - \omega^2[4 - 3(1 - c)\rho] - 3(1 - c)(\omega^2 - \rho)} \right]},
\]

\[
\bar{g}_M = \varepsilon - \frac{(1 + c)\Delta\bar{\ep}_t^2}{4c\Delta\ep} + \frac{(1 - c)\varepsilon\Delta\ep(\omega^2 - 1)}{12c(\omega^2 - 1) + (1 - c)\Delta\ep(\omega^2 - \rho)^2} \times \left[ 1 - \frac{9\omega^2[1 + 3c(3 - 2\rho)^2 - 4c(6 + \rho(3\rho - 8))] + 9[2 - 3(1 - c)\rho] [1 + c(2\rho - 3)]}{5 + 3c(8\rho - 11) - 2\rho}[2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1 - c)(2\omega^2 + 1)] \times (1 - c)\Delta\bar{\ep}_t \right] \times \left[ 1 - \frac{\Delta\bar{\ep}_t [1 + (1 + 3c)(\rho + 3\omega^4 + 1) - (5 + \rho + 3c(4\rho + 1))\omega^2]}{6c(\omega^2 - 1)^2} \right],
\]

(3.53 cont.)
3. A general closed-form solution in the small-deformation limit

\[
\tilde{h}_M = \frac{\varepsilon}{2} - \frac{\Delta \tilde{\varepsilon}^2}{2c^2 \Delta \varepsilon} - \frac{(1 - c) \Delta \tilde{\varepsilon}^2}{24c^2 \varepsilon (\omega^2 - 1)} \left[ (2 + 3c)(2\rho - 1) - 2\rho - \frac{9c(\rho - 1)}{\omega^2 - 1} \right] - \\
\frac{(1 - c) \varepsilon \Delta \tilde{\varepsilon}^2 (4\rho - 1)}{24 \left[ \varepsilon (\omega^2 - 1) + (1 - c) \Delta \varepsilon (\rho - 1) \right]^2} \times \\
\left\{ 1 - \frac{9 \left[ 2 + c(2\rho - 3)\omega^2 - (3 - c)\rho + \omega^2 \right]}{(4\rho - 1) \left[ 2 + (5 - 9c)\omega^2 + 2\omega^4 - 3\rho(1 - c)(2\omega^2 + 1) \right]} \right\}. 
\]

(3.53 cont.)

The sets of expressions (3.51), (3.52), (3.53) correspond to dielectric elastomer composites with particles and elastomers that exhibit limiting constitutive behaviors. The result (3.49) also contains special cases associated with limiting geometries of the particles and their spatial distribution. We spell out two of them in the following sections.

3.5 The case of isotropic distributions of spherical particles with isotropic elastic dielectric properties

The practical motivation to consider isotropic distributions of spherical particles stems from recent experimental findings, including those of Zhang et al. (2002), Huang and Zhang (2004), Huang et al. (2005), Carpi and De Rossi (2005), Mc Carthy et al. (2009), and Liu et al. (2013), which have shown that the addition of random distributions of roughly spherical particles, made up of high-permittivity or (semi-)conducting solids, into dielectric elastomers leads to a drastic enhancement of the electrostrictive capabilities of these materials. Furthermore, we seek to identify what other type of fillers not yet utilized in experimental studies, such as liquid-like particles with high-permittivity and vacuous pores, may potentially lead to the enhancement of the overall elastic dielectric properties of dielectric elastomers.

When the aspect ratio \( \omega = 1 \), the underlying microstructure reduces to an isotropic distribution of spherical particles\(^6\) (see Fig. 3.3(a)) and the thirteen effective constants (3.49) reduce to just five (two elastic, one dielectric, and two electrostrictive) independent effective constants. For completeness, the centers of the spheres are therefore distributed with isotropic symmetry \( \mathbf{Z} = \mathbf{I} \) and the microstructural tensors (3.36) (or indeed (3.42)) reduce to

\[
\langle \xi_i \xi_j \rangle = \frac{1}{3} \delta_{ij} \quad \text{and} \quad \langle \xi_i \xi_j \xi_k \xi_l \rangle = \frac{1}{15} (\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}).
\]

(3.54)
we record here the specialization of the effective electromechanical constants (3.49):

\[
\begin{align*}
\bar{c}_L &= 2\kappa + \frac{2}{3}\mu, \quad \bar{d}_L = \kappa + \frac{4}{3}\mu, \quad \bar{e}_L = \bar{f}_L = 2\mu, \quad \bar{g}_L = \kappa - \frac{2}{3}\mu; \\
\bar{c}_M &= \frac{1}{3}\bar{m}_K + \frac{2}{3}\bar{m}_J, \quad \bar{d}_M = \frac{2}{3}\bar{m}_K + \frac{1}{3}\bar{m}_J, \quad \bar{e}_M = \bar{f}_M = \bar{m}_K, \quad \bar{g}_M = \bar{h}_M = \frac{1}{3}(\bar{m}_J - \bar{m}_K),
\end{align*}
\]

where

\[
\begin{align*}
\bar{\mu} &= \mu + \frac{5c(3\kappa + 4\mu)(\mu_p - \mu)}{[(6c + 9)\kappa + 4(3c + 2)\mu]\mu + 6(1 - c)(\kappa + 2\mu)\mu_p}, \\
\bar{\kappa} &= \kappa + \frac{c(3\kappa + 4\mu)(\kappa_p - \kappa)}{3\kappa_p + 4\mu - 3c(\kappa_p - \kappa)}; \\
\bar{\varepsilon} &= \varepsilon + \frac{3c(\varepsilon_p - \varepsilon)}{(2 + c)\varepsilon + (1 - c)\varepsilon_p}, \\
\bar{\bar{m}}_K &= \varepsilon + \frac{1 - c}{15c\varepsilon} \frac{\Delta \varepsilon^2 + 3(1 - c)(\kappa + 2\mu)\Delta \varepsilon \Delta \bar{\mu} + 1}{\Delta \varepsilon^2 \Delta \bar{\mu}} \left[ \frac{3(1 - c)[\mu((5 - c)\varepsilon + (1 + c)\varepsilon_p) + 3\varepsilon \kappa]}{c^2 \varepsilon \mu (3\kappa + 4\mu)} + \frac{(14 + c)\varepsilon + (1 - c)\varepsilon_p}{c^2 \varepsilon (\mu_p - \mu)} \right], \\
\bar{\bar{m}}_J &= \varepsilon + \frac{3(1 - c)\varepsilon \varepsilon_p (\varepsilon - \varepsilon_p)}{2[(2 + c)\varepsilon + (1 - c)\varepsilon_p]^2[3c(\kappa - \kappa_p) + 3\kappa_p + 4\mu]} + \frac{3c\varepsilon^2 (\varepsilon - \varepsilon_p)[3((c - 4)c + 6)\kappa - 3(c - 4)(c - 1)\kappa_p + 4(2 + c)\mu]}{2[(2 + c)\varepsilon + (1 - c)\varepsilon_p]^2[3c(\kappa - \kappa_p) + 3\kappa_p + 4\mu]} \right],
\end{align*}
\]

with \(\kappa = \lambda + 2\mu/3, \kappa_p = \lambda_p + 2\mu_p/3, \Delta \bar{\mu} = \bar{\mu} - \mu, \Delta \bar{\varepsilon} = \bar{\varepsilon} - \varepsilon\). In these expressions, \(\mu, \mu_p, \kappa, \kappa_p, \varepsilon, \varepsilon_p\) stand for the shear modulus, bulk modulus, and permittivity of the matrix and the particles, while \(\bar{\mu}, \bar{\kappa}, \bar{\varepsilon}, \bar{\bar{m}}_K, \) and \(\bar{\bar{m}}_J\) denote respectively the effective shear modulus, bulk modulus, permittivity, and electrostrictive coefficients of the composite.

The solutions (3.29), (3.30), (3.31) therefore specialize for such a class of isotropic dielectric elastomer composite to

\[
\bar{\mathbf{L}} = 2\bar{\mu} \mathbf{K} + 3\bar{\kappa} \mathbf{J}, \quad \bar{\varepsilon} = \bar{\varepsilon} \mathbf{I}, \quad \bar{\mathbf{M}} = \bar{\bar{m}}_K \mathbf{K} + \bar{\bar{m}}_J \mathbf{J},
\]

where it is recalled that the orthogonal projections tensor \(\mathbf{K}\) and \(\mathbf{J}\) are given by expressions (3.20). The overall electromechanical constitutive response (2.26)-(2.27) for this type of isotropic dielectric
elastomer composites read then simply as

\[
\mathbf{S} = \tilde{L} \mathbf{H} + \tilde{M} \mathbf{E} \otimes \mathbf{E}
\]

\[
= (2\tilde{\mu} \mathbf{K} + 3\tilde{\kappa} \mathbf{J}) \mathbf{H} + (\tilde{m}_K \mathbf{K} + \tilde{m}_J \mathbf{J}) \mathbf{E} \otimes \mathbf{E}
\]

\[
= \tilde{\mu} \left[ \mathbf{H} + \mathbf{H}^T - \frac{2}{3}(\text{tr} \mathbf{H}) \mathbf{I} \right] + \tilde{\kappa}(\text{tr} \mathbf{H}) \mathbf{I} + \tilde{m}_K \left[ \mathbf{E} \otimes \mathbf{E} - \frac{1}{3} (\mathbf{E} \cdot \mathbf{E}) \mathbf{I} \right] + \frac{\tilde{m}_J}{3} (\mathbf{E} \cdot \mathbf{E}) \mathbf{I}
\]

(3.58)

and

\[
\mathbf{D} = \tilde{\varepsilon} \mathbf{E} = \bar{\varepsilon} \mathbf{E}
\]

(3.59)

to leading order in the limit of small deformations and moderate electric fields.

Again, the above results contain several limiting cases worth recording explicitly:

- **Dilute volume fraction of particles.** In the fundamental limit when the particles are present in dilute volume fraction as \( c \to 0^+ \), the effective electromechanical material parameters (3.56) reduce asymptotically to

\[
\tilde{\mu} = \mu + \frac{5(3\kappa + 4\mu)(\mu_p - \mu)}{(9\kappa + 8\mu)\mu + 6(\kappa + 2\mu)\mu_p} c,
\]

\[
\tilde{\kappa} = \kappa + \frac{(3\kappa + 4\mu)(\kappa_p - \kappa)}{3\kappa_p + 4\mu} c,
\]

\[
\tilde{\varepsilon} = \varepsilon + \frac{3(\varepsilon_p - \varepsilon)}{2(\varepsilon + \varepsilon_p)} c
\]

\[
\tilde{m}_K = \varepsilon + \frac{3(\varepsilon_p - \varepsilon)(9\kappa - 6\kappa_p + 4\mu)}{2(\varepsilon + \varepsilon_p)(3\kappa_p + 4\mu)} c
\]

\[
\tilde{m}_J = -\frac{\varepsilon}{2} + \frac{3(\varepsilon_p - \varepsilon)(9\kappa - 6\kappa_p + 4\mu)}{2(\varepsilon + \varepsilon_p)(3\kappa_p + 4\mu)} c
\]

(3.60)

up to \( O(c) \).

- **Rigid particles with infinite permittivity.** In the limit of rigid infinite-permittivity particles when \( \mu_p, \kappa_p = +\infty \) and \( \varepsilon_p = +\infty \), the effective electromechanical material parameters (3.56) reduce to

\[
\tilde{\mu} = \mu + \frac{5c(3\kappa + 4\mu)}{6(1-c)(\kappa + 2\mu)} \mu,
\]

\[
\tilde{\kappa} = \kappa + \frac{c(3\kappa + 4\mu)}{3(1-c)} c,
\]

\[
\tilde{\varepsilon} = \varepsilon + \frac{3c}{1-c} \varepsilon
\]

\[
\tilde{m}_K = \varepsilon + \frac{3c(1-c)}{10(1-c)^2(\kappa + 2\mu)} \varepsilon,
\]

\[
\tilde{m}_J = -\frac{\varepsilon}{2} + \frac{3c(2+c)}{2(1-c)^2} \varepsilon.
\]

(3.61)
If, in addition, the underlying elastomeric matrix is incompressible \( (\kappa = +\infty) \), these formulae reduce further to

\[
\begin{align*}
\bar{\mu} &= \mu + \frac{5c}{2(1-c)} \mu, \quad \bar{\kappa} = +\infty, \quad \bar{\varepsilon} = \varepsilon + \frac{3c}{1-c} \varepsilon, \\
\bar{m}_K &= \varepsilon + \frac{21c}{10(1-c)} \varepsilon, \quad \bar{m}_J = -\frac{\varepsilon}{2} + \frac{3c(2+c)}{2(1-c)^2} \varepsilon.
\end{align*}
\] (3.62)

The results (3.61)–(3.62) are relevant for dielectric elastomer composites wherein the filler particles are typical ceramics (e.g., titania) or metals (e.g., iron), which generally exhibit much larger stiffness and permittivity (infinitely larger for the case of metals) than elastomers.

- **Liquid particles with infinite permittivity.** The case \( \mu_p = 0, \kappa_p = +\infty, \) and \( \varepsilon_p = +\infty \) corresponds to particles that are liquid-like (incompressible with vanishingly small shear modulus) and of infinite permittivity. Granted these limiting values for the properties of the particles, the effective electromechanical material parameters (3.56) reduce to

\[
\begin{align*}
\bar{\mu} &= \mu - \frac{5c(3\kappa + 4\mu)}{(9 + 6c)\kappa + 4(2 + 3c)\mu} \mu, \quad \bar{\kappa} = \kappa + \frac{c(3\kappa + 4\mu)}{3(1-c)} \mu, \quad \bar{\varepsilon} = \varepsilon + \frac{3c}{1-c} \varepsilon, \\
\bar{m}_K &= \varepsilon + \frac{3c[3(7c + 3)\kappa + (27c - 17)\mu]}{5(1-c)[(9 + 6c)\kappa + 4(2 + 3c)\mu]} \varepsilon, \quad \bar{m}_J = -\frac{\varepsilon}{2} + \frac{3c(2+c)}{2(1-c)^2} \varepsilon.
\end{align*}
\] (3.63)

When the underlying elastomeric matrix is incompressible \( (\kappa = +\infty) \), these formulae reduce further to

\[
\begin{align*}
\bar{\mu} &= \mu - \frac{5c}{3 + 2c} \mu, \quad \bar{\kappa} = +\infty, \quad \bar{\varepsilon} = \varepsilon + \frac{3c}{1-c} \varepsilon, \\
\bar{m}_K &= \varepsilon + \frac{3c(3 + 7c)}{5(1-c)(3 + 2c)} \varepsilon, \quad \bar{m}_J = -\frac{\varepsilon}{2} + \frac{3c(2+c)}{2(1-c)^2} \varepsilon.
\end{align*}
\] (3.64)

Compared with elastomers, many fluids (e.g., water) and eutectic alloys (e.g., Galinstan) can be idealized as incompressible and as having zero shear modulus and infinite permittivity (see, e.g., Wang et al., 2012; Fassler and Majidi, 2015). Expressions (3.63)–(3.64) are relevant for such type of filler particles.

- **Porous dielectric elastomers.** The limiting values \( \mu_p, \kappa_p, \varepsilon_p = 0, \kappa = +\infty, \) where \( \varepsilon_0 \) stands for the permittivity of vacuum, correspond to an incompressible matrix containing an isotropic distribution of vacuous pores. In this case, the effective electromechanical material
parameters (3.56) reduce to

\[
\begin{align*}
\bar{\mu} &= \mu - \frac{5c}{3 + 2e} \mu, \\
\bar{\kappa} &= \frac{4(1 - c)}{3c} \mu, \\
\bar{\varepsilon} &= \varepsilon + \frac{3c(\varepsilon_0 - \varepsilon)\varepsilon}{(2 + c)\varepsilon + (1 - c)\varepsilon_0}, \\
\bar{m}_K &= \varepsilon - \frac{3c(\varepsilon - \varepsilon_0)[((26 + 7c)c + 42)\varepsilon + ((4 - 7c)c + 3)\varepsilon_0]}{5[3 + 2c][(2 + c)\varepsilon + (1 - c)\varepsilon_0]^2} \varepsilon, \\
\bar{m}_J &= -\frac{\varepsilon}{2} + \frac{3(\varepsilon - \varepsilon_0)[((c - 4)c + 6)\varepsilon - (c - 1)(3 + c)\varepsilon_0]}{2[(2 + c)\varepsilon + (1 - c)\varepsilon_0]^2} \varepsilon.
\end{align*}
\] (3.65)

3.6 The case of transversely isotropic distributions of aligned cylindrical fibers with circular cross section and isotropic elastic dielectric properties

A number of studies (see, e.g., Wang and Mark, 1990; Meddeb and Ounaies, 2012; Lu et al., 2012) have indicated that anisotropic fillers in the form of short (needle-like) and long (cylindrical) fibers can potentially lead to even larger enhancements than those endowed by spherical fillers. In addition, such a type of composites can be readily fabricated by means of modern synthesis/manufacturing processes (see, e.g., Lu et al. (2012); Park et al. (2012); López Jiménez and Pellegrino (2012)). Finally, we seek to identify what type of fillers may potentially lead to the enhancement of the overall elastic dielectric properties of dielectric elastomers. This is achieved by recording results for mechanically stiff fibers, liquid-like fibers with high-permittivity or vacuous cylindrical pores.

When the aspect ratio \( \omega = \infty \), the underlying microstructure reduces to a transversely isotropic distribution of cylindrical fibers with circular cross section (see Fig. 3.3(b)). In this limiting case,
expressions (3.49) simplify to

\[ \bar{c}_L = 2\lambda + 2\mu + \frac{2c(\lambda + 2\mu)(\Delta \lambda + \Delta \mu)}{\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)}, \]

\[ \bar{d}_L = \lambda + 2\mu + c(\Delta \lambda + 2\Delta \mu) - \frac{c(1 - c)\Delta \lambda^2}{\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)}, \]

\[ \bar{e}_L = 2\mu + \frac{4c\mu \Delta \mu (\lambda + 2\mu)}{2\mu(\lambda + 2\mu) + (1 - c)\Delta \mu (\lambda + 3\mu)}; \]

\[ \bar{f}_L = 2\mu + \frac{4c\mu \Delta \mu}{2\mu + (1 - c)\Delta \mu}, \]

\[ \bar{g}_L = \lambda + \frac{c\Delta \lambda (\lambda + 2\mu)}{\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)}; \]

\[ \bar{e}_t = \varepsilon + \frac{2c\varepsilon \Delta \varepsilon}{2\varepsilon + (1 - c)\Delta \varepsilon}; \]

\[ \bar{e}_t = \varepsilon + c\Delta \varepsilon; \]

\[ \bar{v}_M = \varepsilon + \frac{2c(1 - c)\varepsilon \Delta \varepsilon (2\varepsilon + \Delta \varepsilon)(\Delta \lambda + \Delta \mu)}{[2\varepsilon + (1 - c)\Delta \varepsilon]^2[\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)],} \]

\[ \bar{d}_M = \varepsilon + \frac{c\Delta \varepsilon}{2} \left[ 1 + \frac{(1 - c)\Delta \lambda}{\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)} \right], \]

\[ \bar{e}_M = \varepsilon + \frac{4c\varepsilon^2 \Delta \varepsilon}{[2\varepsilon + (1 - c)\Delta \varepsilon]^2} \left[ 1 + \frac{(1 - c)\Delta \varepsilon (4\mu(\lambda + 2\mu) + 3(1 - c)\lambda \Delta \mu + (11 - 7c)\mu \Delta \mu)}{8\varepsilon[2\mu(\lambda + 2\mu) + (1 - c)\Delta \mu (\lambda + 3\mu)]} \right], \]

\[ \bar{f}_M = \varepsilon + \frac{2c\varepsilon \Delta \varepsilon}{2\varepsilon + (1 - c)\Delta \varepsilon}, \]

\[ \bar{g}_M = -\frac{\varepsilon}{2} - \frac{c\varepsilon \Delta \varepsilon}{2\varepsilon + (1 - c)\Delta \varepsilon} + \frac{c(1 - c)\varepsilon \Delta \varepsilon (2\varepsilon + \Delta \varepsilon) \Delta \lambda}{[2\varepsilon + (1 - c)\Delta \varepsilon]^2[\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)]}, \]

\[ \bar{h}_M = -\frac{\varepsilon}{2} - \frac{c\Delta \varepsilon (\lambda + 2\mu)}{2[\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)]}, \]

\[ (3.66) \]

where it is recalled that \( \Delta \mu = \mu_p - \mu, \Delta \lambda = \lambda_p - \lambda, \Delta \varepsilon = \varepsilon_p - \varepsilon. \)
Analytical solutions for differential coated microstructures in the small-deformation limit


In this chapter, we consider another special class of microstructures that allow to construct rigorous analytical solutions for the effective electromechanical tensors $\tilde{\mathbf{L}}$, $\tilde{\mathbf{\epsilon}}$, and $\tilde{\mathbf{M}}$ that characterize the response in the limit of small deformations and moderate electric fields: (i) a differential coated sphere assemblage corresponding to an isotropic suspension of spherical particles and (ii) a differential coated cylinder assemblage corresponding to a transversely isotropic distribution of aligned fibers with circular cross-section. A defining feature of these microstructures is that the boundary value problems (2.36) and (2.37) for the tensors $\Gamma$ and $\gamma$ — needed in the computation of the effective electromechanical tensors (2.33)-(2.35) — over the entire composite need only be solved over a single coated sphere and a single coated cylinder.
4.1 The overall electromechanical response of a differential coated sphere assemblage

In this section, we derive the analytical solution for the electromechanical response of a differential coated sphere assemblage (DCS) in the limit of small deformations and moderate electric fields. We restrict attention in this section to such isotropic suspensions of spherical particles wherein the matrix and the particles are isotropic elastic dielectric materials. It follows that the tensors $L^{(r)}, \epsilon^{(r)}, M^{(r)}$ defined by (2.31) are of the form

\[
L^{(1)} = 2\mu K + 3\kappa J, \quad \epsilon^{(1)} = \epsilon I, \quad M^{(1)} = m_K K + m_J J,
\]

\[
L^{(2)} = 2\mu p K + 3\kappa p J, \quad \epsilon^{(2)} = \epsilon_p I, \quad M^{(2)} = m_{K_p} K + m_{J_p} J.
\] (4.1)

We recall that in these expressions, $\mu, \mu_p, \kappa, \kappa_p, \epsilon, \epsilon_p, m_K, m_K_p, m_J, m_J_p$ stand for, respectively, the shear modulus, bulk modulus, permittivity, and electrostrictive coefficients of the matrix and the particles, and $K, J$ are two orthogonal projection tensors given by (3.20). Moreover, because of the assumed overall (geometric and constitutive) isotropy of the suspension, the effective tensors (2.33)–(2.35) are also of the form

\[
\tilde{L} = 2\tilde{\mu} K + 3\tilde{\kappa} J, \quad \tilde{\epsilon} = \tilde{\epsilon} I, \quad \tilde{M} = \tilde{m}_K K + \tilde{m}_J J,
\] (4.2)

much like in Section 3.5.

We recall that a coated sphere assemblage (Hashin, 1962) is a two-phase particulate microstructure wherein homothetic coated spheres — comprising a spherical core made up of the particle material that is surrounded by a spherical shell made up of the matrix material — of infinitely many sizes are assembled together to fill the entire domain $\Omega$ occupied by the composite. The particular manner in which the coated spheres are assembled is arbitrary. For assemblages wherein coated spheres of comparable size are placed far apart from each other and surrounded by coated spheres of much smaller size, in such a way that the microstructure is fractal-like comprising a hierarchy of well-separated coated spheres of infinitely many sizes, it is possible to construct analytical solutions for the corresponding boundary value problems (2.36) and (2.37). Following the terminology of Avellaneda (1987) (see also the prior work of Milton (1985) and Section 10.5 in Milton, 2002), we refer to such assemblages as differential coated sphere assemblages. One of two defining features of such microstructures is that, by construction, any coated sphere in the assemblage can be regarded to be surrounded by a homogeneous medium of infinite extent with the effective
properties of the entire assemblage. The other defining feature being that the average response of any coated sphere is the same as the average response of the entire assemblage (Milton, 1985; Milton, 2002) (or, equivalently, that the so-called average “polarizability” of each coated sphere vanishes (Milton, 1985)).

For the elastostatics and electrostatics problems (2.36) and (2.37) of interest here, the above-outlined features of a differential coated sphere assemblage entail that the gradients of the fields $\Gamma$ and $\gamma$ are the same in each one of the coated spheres and, therefore, that the formulae (2.33)–(2.35) for the effective electromechanical tensors $\tilde{L}$, $\tilde{\epsilon}$, $\tilde{M}$ reduce to

\[
\tilde{L}_{ijkl} = \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} L_{i,jrs} \Gamma_{rkl,s} \, dX, \tag{4.3}
\]
\[
\tilde{\epsilon}_{ij} = \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} \epsilon_{is} \gamma_{s,j} \, dX, \tag{4.4}
\]
\[
\tilde{M}_{ijkl} = \frac{1}{|\mathcal{B}|} \int_{\mathcal{B}} \Gamma_{rij,s} M_{rsuv} \gamma_{u,k} \gamma_{v,l} \, dX, \tag{4.5}
\]

where the integrals are now over a single coated sphere, denoted as $\mathcal{B}$, as opposed to over the entire assemblage $\Omega$. Moreover, the gradients of the fields $\Gamma$ and $\gamma$ needed in (4.3)–(4.5) can be expediently computed by taking the domain $\Omega$ in the boundary value problems (2.36) and (2.37) to be an infinite body comprised of a single coated sphere, occupying the domain $\mathcal{B} = \{X : |X| \leq 1\}$ say, embedded in a homogeneous medium with the effective properties of the entire assemblage. Namely, for the elastostatics problem (2.36) it suffices to consider $\Omega = \mathbb{R}^3$ and the local modulus of elasticity

\[
L = \begin{cases} 
L^{(2)} = 2\mu_p \mathcal{K} + 3\kappa_p \mathcal{J} & \text{if } |X| \leq R_p \\
L^{(1)} = 2\mu \mathcal{K} + 3\kappa \mathcal{J} & \text{if } R_p < |X| \leq 1, \\
\bar{L} = 2\bar{\mu} \mathcal{K} + 3\bar{\kappa} \mathcal{J} & \text{if } |X| > 1
\end{cases} \tag{4.6}
\]

while for the electrostatics problem (2.37) it suffices to consider $\Omega = \mathbb{R}^3$ and the local permittivity

\[
\epsilon = \begin{cases} 
\epsilon^{(2)} = \varepsilon_p \mathcal{I} & \text{if } |X| \leq R_p \\
\epsilon^{(1)} = \varepsilon \mathcal{I} & \text{if } R_p < |X| \leq 1, \\
\bar{\epsilon} = \bar{\varepsilon} \mathcal{I} & \text{if } |X| > 1
\end{cases} \tag{4.7}
\]

where $R_p$ stands for the radius of the particle within the coated sphere of radius 1. We note for later reference that the local electrostrictive tensor is given by

\[
M = \begin{cases} 
M^{(2)} = m_K \mathcal{K} + m_J \mathcal{J} & \text{if } |X| \leq R_p \mathcal{J} \\
M^{(1)} = m_K \mathcal{K} + m_J \mathcal{J} & \text{if } R_p < |X| \leq 1, \\
\tilde{M} = \tilde{m}_K \mathcal{K} + \tilde{m}_J \mathcal{J} & \text{if } |X| > 1
\end{cases} \tag{4.8}
\]
4. Analytical solutions for differential coated microstructures in the small-deformation limit

Given (4.6) and (4.7), the solutions to the boundary value problems (2.36) and (2.37) can be readily worked out in terms of solid harmonics. Before proceeding with the presentation of the solutions, we remark that

\[ \tilde{\mu} = \tilde{L}_{1212} = \frac{1}{|B|} \int_B L_{12r,s} \Gamma_{r,s} dX, \]

\[ \tilde{\kappa} = \frac{1}{9} \tilde{L}_{ii} = \frac{1}{|B|} \int_B \frac{1}{9} L_{iirs} \Gamma_{r,s} dX, \]

\[ \tilde{\varepsilon} = \frac{1}{3} \tilde{\varepsilon} = \frac{1}{|B|} \int_B \frac{1}{3} \tilde{\varepsilon}_{i} dX, \]

\[ \tilde{m}_K = 2 \tilde{M}_{1212} = \frac{1}{|B|} \int_B 2 \Gamma_{r,s} M_{rsuv} \gamma_{u,1} \gamma_{v,2} dX, \]

\[ \tilde{m}_J = \frac{1}{3} \tilde{M}_{ij} = \frac{1}{|B|} \int_B \frac{1}{3} \Gamma_{rii,s} M_{rsuv} \gamma_{u,j} \gamma_{v,j} dX. \]

That is, by virtue of the isotropic form (4.2) of the effective tensors \( \tilde{L}, \tilde{\varepsilon}, \tilde{M} \), only certain combinations of the components of the gradients of the fields \( \Gamma \) and \( \gamma \) are needed for their computation. In the sequel, for conciseness, we report solutions for such combinations.

4.1.1 The solution for \( \Gamma \)

We begin by presenting the solution for the components \( \Gamma_{i12,j} \), which are needed in the computation of the effective shear modulus (4.9) and the effective electrostrictive coefficient (4.12). Physically, such components are associated with the elastic response of the suspension under simple shear loading. As alluded to above, the solution can be easily constructed in terms of solid harmonics (see, e.g., Chapter XI in Love, 1906). It reads as follows:

\[ \Gamma_{112,1} = \left[ \frac{f'}{R} + 2g \right] X_1 X_2 + \frac{g'}{R} X_1^2 X_2, \]

\[ \Gamma_{212,2} = \left[ \frac{f'}{R} + 2g \right] X_2 X_2 + \frac{g'}{R} X_1 X_2^2, \]

\[ \Gamma_{312,3} = g X_1 X_2 + \frac{g'}{R} X_1 X_2 X_3^2, \]

\[ \Gamma_{112,2} = \Gamma_{212,1} = \frac{1}{2} \left[ \frac{f'}{R} + g \right] (X_1^2 + X_2^2) + f + \frac{g'}{R} X_1^2 X_2^2, \]

\[ \Gamma_{112,3} = \Gamma_{312,1} = \frac{1}{2} \left[ \frac{f'}{R} + g \right] X_2 X_3 + \frac{g'}{R} X_1 X_2^2 X_3, \]

\[ \Gamma_{212,3} = \Gamma_{312,2} = \frac{1}{2} \left[ \frac{f'}{R} + g \right] X_1 X_3 + \frac{g'}{R} X_1 X_2 X_3, \]
where \( f \) and \( g \) are functions of \( R = |X| \) given by

\[
f = \begin{cases} 
A_1 + A_3 R^2 & \text{if } 0 \leq R \leq R_p \\
B_1 + \frac{B_2}{R^5} + B_3 R^2 + \frac{B_4}{R^3} & \text{if } R_p < R \leq 1
\end{cases}
\]

and

\[
g = \begin{cases} 
-\frac{2(6\kappa_p + 17\mu_p)}{15\kappa_p + 11\mu_p} A_3 & \text{if } 0 \leq R \leq R_p \\
-\frac{5B_2}{R^3} - \frac{2(6\kappa + 17\mu)}{15\kappa + 11\mu} B_3 + \left[1 + \frac{3\mu}{\mu} \right] \frac{B_4}{R^5} & \text{if } R_p < R \leq 1
\end{cases}
\]

In these expressions, use has been made of the notation \( f'(R) = df(R)/dR \), \( g'(R) = dg(R)/dR \), and \( A_1, A_3, B_1, B_2, B_3, B_4 \) are constants that depend on the shear and bulk moduli of the particles, \( \mu_p, \kappa_p \), the matrix, \( \mu, \kappa \), the suspension, \( \tilde{\mu}, \tilde{\kappa} \), as well as on the concentration of particles, \( c = R_p^3 \).

Next, we present the solution for the combination of components \( \Gamma_{i k k,j} \). This combination is needed in the computation of the effective bulk modulus (4.10) and the effective electrostrictive coefficient (4.13). Physically, it characterizes the elastic response of the suspension under hydrostatic loading. It can be compactly written as follows:

\[
\Gamma_{i k k,j} = \left[ d + \frac{e}{R^3} \right] \delta_{ij} - \frac{3e}{R^3} X_i X_j,
\]

where

\[
d = \begin{cases} 
\frac{(3\kappa + 4\mu)(3\tilde{\kappa} + 4\tilde{\mu})}{12c(\kappa_p - \kappa)(\mu - \tilde{\mu}) + (3\kappa + 4\mu)(3\kappa_p + 4\mu)} & \text{if } 0 \leq R \leq R_p \\
\frac{(3\kappa_p + 4\mu)(3\tilde{\kappa} + 4\tilde{\mu})}{12c(\kappa_p - \kappa)(\mu - \tilde{\mu}) + (3\kappa + 4\mu)(3\kappa_p + 4\mu)} & \text{if } R_p < R \leq 1
\end{cases}
\]

and

\[
e = \begin{cases} 
0 & \text{if } 0 \leq R \leq R_p \\
\frac{3(\kappa - \kappa_p)(3\tilde{\kappa} + 4\tilde{\mu})}{12c(\kappa_p - \kappa)(\mu - \tilde{\mu}) + (3\kappa + 4\mu)(3\kappa_p + 4\mu)} & \text{if } R_p < R \leq 1
\end{cases}
\]

**4.1.2 The solution for \( \gamma \)**

Finally, we present the solution for the gradient \( \gamma_{i,j} \), which is needed in the computation of the effective permittivity (4.11), as well as in the computation of the two effective electrostrictive coefficients (4.12) and (4.13). Physically, it characterizes the dielectric response of the suspension under an arbitrary affine electric field. The solution is given by

\[
\gamma_{i,j} = - \left[ a + \frac{b}{R^3} \right] \delta_{ij} + \frac{3b}{R^3} X_i X_j,
\]
explicit solutions for equations turns out to be physical. On the other hand, expressions (4.12) and (4.13) directly render (4.9), (4.10), (4.11) render polynomial equations for such effective constants. Only one root of these results (4.14), (4.17), (4.20) for the local fields $\Gamma_i$ that characterize the overall elastic dielectric response of the suspension. Making use of the electromechanical constants can be written as the integrals (4.9) through (4.13). Since the fields $\Gamma_i$ that depend on $\tilde{\mu}$, $\tilde{\kappa}$, $\tilde{\varepsilon}$, expressions (4.9), (4.10), (4.11) render polynomial equations for such effective constants. Only one root of these equations turns out to be physical. On the other hand, expressions (4.12) and (4.13) directly render explicit solutions for $\tilde{m}_K$ and $\tilde{m}_J$. After some algebraic manipulation, the result for all five effective electromechanical constants can be written as

\[
\tilde{\mu}^{DCS} = \frac{q_2 + \sqrt{q_2^2 + 4q_1q_3}}{2q_1} \mu, \\
\tilde{\kappa}^{DCS} = \kappa + \frac{c(3\kappa + 4\mu)(\kappa_p - \kappa)}{3\kappa_p + 4\mu - 3c(\kappa_p - \kappa)}, \\
\tilde{\varepsilon}^{DCS} = \varepsilon + \frac{3c(\varepsilon_p - \varepsilon)\varepsilon}{(2 + c)\varepsilon + (1 - c)\varepsilon_p}, \\
\tilde{m}_K^{DCS} = 2A_1a_p^2m_K\mu + \frac{42A_2a_p^2\varepsilon^{5/3}(3\kappa_p + \mu_p)m_{K_p}}{75\kappa_p + 55\mu_p} + \frac{2}{5}B_1 \left( \frac{1}{c} - 1 \right) \left( 5a_p^2c + b_p^2 \right) m_K - \frac{27}{5}B_2b_p^2 \left( 1 - \frac{1}{c^{8/3}} \right) m_K - \frac{252B_3b_p^2 \left( a_p^2c + b_p - c^{1/3}(a_p + b_p) \right) m_J}{5c^{1/3}(15\kappa + 11\mu)} + \frac{6B_3b_p^2(1 - c^{1/3})m_K}{5c^{1/3}} + \frac{42B_3a_p \left( a_p \left( c^{1/3} - c^2 \right)(3\kappa + \mu) + b_n \left( c - c^{1/3} \right)(3\kappa - 2\mu) \right) m_K}{5c^{1/3}(15\kappa + 11\mu)} - \frac{B_4b_p(1 - c)\left( b_n(1 + c)(15\kappa + 4\mu) - 2a_p^2c(3\kappa + 8\mu) \right) m_K}{5c^{1/3}(15\kappa + 11\mu)} - \frac{2B_4b_p(1 - c)\left( b_n - 4a_p^2c + b_n \right) m_J}{5c^2}, \\
\tilde{m}_J^{DCS} = \left( 1 - c \right) (\tilde{\varepsilon} - \varepsilon)^2(\tilde{\kappa} - \kappa) \left[ \frac{(3\kappa_p + 4\mu) \left( 2(2 + c)\varepsilon^2 + 4(1 - c)\varepsilon \varepsilon_p + (1 + 2c)\varepsilon_p^2 \right) m_J}{9c^2(\varepsilon_p - \varepsilon)^2(\kappa_p - \kappa)(3\kappa + 4\mu)} + \frac{[(7 - c)\varepsilon + (5 + c)\varepsilon_p] m_K}{3c^2(\varepsilon_p - \varepsilon)(3\kappa + 4\mu)} + \left( \tilde{\varepsilon} - \varepsilon \right)^2(\tilde{\kappa} - \kappa) m_J \right] + \frac{(\tilde{\varepsilon} - \varepsilon)^2(\tilde{\kappa} - \kappa) m_J}{c^2(\varepsilon_p - \varepsilon)^2(\kappa_p - \kappa)}.
\]
where the superscript “DCS” has been appended for clarity and later reference. It is however dropped when unnecessary or bulky in the remaining of this section for notational simplicity. The variables \( q_1, q_2, q_3 \) in the above formulae are given by expressions (B.2) in Appendix B.1.2, and it is recalled that \( A_1, A_3, B_1, B_2, B_3, B_4 \) are defined in Appendix B.1.1, while \( a_p, a_n, b_n \) are defined in expressions (4.21) and (4.22). A number of theoretical and practical remarks are of note:

\( \text{i.} \) The solution (4.23) is valid for any choice of the shear moduli \( \mu, \mu_p, \) bulk moduli \( \kappa, \kappa_p, \) permittivities \( \varepsilon, \varepsilon_p, \) and electrostrictive coefficients \( m_K, m_K', m_J, m_J' \) characterizing the elastic dielectric response of the underlying rubber matrix and the particles, as well as for any choice of the concentration of particles \( c \in [0, 1] \).

\( \text{ii.} \) There are several limiting cases contained in (4.23) worth exploring in detail. In this section, we restrict ourselves to recording explicitly two of them. In the fundamental limit when the particles are present in dilute concentration as \( c \to 0^+ \), the effective constants (4.23) reduce asymptotically to

\[
\begin{align*}
\tilde{\mu}^\text{dil} & = \mu + \frac{5(3\kappa + 4\mu)(\mu_p - \mu)\mu}{(9\kappa + 8\mu)\mu + 6(\kappa + 2\mu)\mu_p} c, \\
\tilde{\kappa}^\text{dil} & = \kappa + \frac{(3\kappa + 4\mu)(\kappa_p - \kappa)}{3\kappa_p + 4\mu} c, \\
\tilde{\varepsilon}^\text{dil} & = \varepsilon + \frac{3(\varepsilon_p - \varepsilon)\varepsilon}{2\varepsilon + \varepsilon_p} c, \\
\tilde{m}_K & = m_K + \left[ \frac{\mu_p(\varepsilon_p - \varepsilon)[36\varepsilon\kappa + 79\varepsilon\mu + 18\varepsilon_p\kappa + 41\varepsilon_p\mu]m_K}{(2\varepsilon + \varepsilon_p)^2[6\mu_p(\kappa + 2\mu) + \mu(9\kappa + 8\mu)]} + \frac{45\varepsilon^2\mu(3\kappa + 4\mu)m_K'}{(2\varepsilon + \varepsilon_p)^2[6\mu_p(\kappa + 2\mu) + \mu(9\kappa + 8\mu)]} \right] c, \\
\tilde{m}_J & = m_J + \left[ \frac{9\varepsilon^2(3\kappa + 4\mu)m_J}{(2\varepsilon + \varepsilon_p)^2(3\kappa_p + 4\mu)} + \frac{3(\varepsilon_p - \varepsilon)(7\varepsilon + 5\varepsilon_p)(\kappa_p - \kappa)m_K}{(2\varepsilon + \varepsilon_p)^2(3\kappa_p + 4\mu)} \right] c,
\end{align*}
\]

(4.24)
to \( O(c) \). These results constitute a generalization of the classical results of Eshelby (1957) and Maxwell (1873) for the purely elastic and purely dielectric overall response of a dilute suspension of spherical particles to the coupled and nonlinear realm of elastic dielectric properties.
iii. When synthesized under typical conditions, rubber can be regarded to be incompressible (Bridgman, 1945), so that $\kappa = +\infty$, and not to exhibit “material” electrostriction (Di Lillo et al., 2011), so that $m_K = -2m_J = \varepsilon$. When compared with rubber, furthermore, typical filler particles such as ceramics and metals can be regarded to be mechanically rigid and also not to exhibit “material” electrostriction, so that $\mu_p, \kappa_p = +\infty$ and $m_{K_p} = -2m_{J_p} = \varepsilon_p$. For this practically relevant choice of rubber and particle properties, the effective constants (4.23) reduce to the more compact form

\begin{equation}
\tilde{\mu}_{\text{rig, inc}} = \mu + \frac{35c\mu}{7 - 15c + 8c^{10/3} + \sqrt{49 + c \left[ 14 + c \left( 8c^{2/3} - 2c^{5/3} - 161c^{2/3} + 294 \right) - 1175 \right]}} ,
\end{equation}

\begin{equation}
\tilde{\kappa}_{\text{rig, inc}} = +\infty ,
\end{equation}

\begin{equation}
\tilde{\varepsilon}_{\text{rig, inc}} = \varepsilon + \frac{3c(\varepsilon_p - \varepsilon)}{(2 + c)(1 - c)} ,
\end{equation}

\begin{equation}
\tilde{m}_{K_{\text{rig, inc}}} = \varepsilon + (\tilde{\varepsilon}_{\text{rig, inc}} - \varepsilon)^2 \left[ \frac{\mu}{8 \left( 1 - c^{2/3} \right) \varepsilon (\tilde{\mu}_{\text{rig, inc}} - \mu)} + \frac{1}{60 \left( 1 + c^{1/3} \right) c \varepsilon} \right] ,
\end{equation}

\begin{equation}
\tilde{m}_{J_{\text{rig, inc}}} = -\varepsilon + \frac{3c(\varepsilon_p - \varepsilon)}{2(2 + c)} \left[ (2 + c)\varepsilon_p + (4 - c)\varepsilon \right] \varepsilon_p^2 ,
\end{equation}

where use has been made of the inequality $\kappa_p \gg \kappa$, typical of standard ceramic and metallic filler particles when compared to rubber, in the computation of the electrostrictive coefficient (4.25).5.

iv. As expected, the effective shear modulus (4.23)\textsubscript{1} agrees identically with the result originally derived by Christensen and Lo (1979), and later proved to be realizable by Avellaneda (1987), for a differential coated sphere assemblage. Further, the effective bulk modulus (4.23)\textsubscript{2} and effective permittivity (4.23)\textsubscript{3} agree identically with the classical results of Hashin (1962) and Hashin and Shtrikman (1962b) for arbitrary (not necessarily differential) coated sphere assemblages. As a corollary, the results (4.23)\textsubscript{2} and (4.23)\textsubscript{3} also agree with one of the Hashin-Shtrikman bounds for the bulk modulus and permittivity of two-phase composites with isotropic (not necessarily particulate) microstructures when the elastic and dielectric properties of the matrix and particles are well ordered.
4. Analytical solutions for differential coated microstructures in the small-deformation limit

v. Similar to the results \((4.23)_2\) and \((4.23)_3\), the result \((4.23)_5\) for the effective electrostrictive coefficient \(\tilde{m}_J\) can be shown (via a neutral inclusion argument) to be exact not just for a differential coated sphere assemblage but for any coated sphere assemblage. This is not true for the result \((4.23)_4\) for the electrostrictive coefficient \(\tilde{m}_K\), which, similar to the effective shear modulus \((4.23)_1\), is exact only for a differential coated sphere assemblage.

vi. Remarkably, the results \((3.56)_2\), \((3.56)_3\), and \((3.56)_5\) for the effective bulk modulus \(\tilde{\kappa}\), effective permittivity \(\tilde{\varepsilon}\), and effective electrostrictive coefficient \(\tilde{m}_J\) of the iterative microstructure considered in Chapter 3 are seen to agree identically with the corresponding effective material parameters \((4.23)_2\), \((4.23)_3\), and \((4.23)_5\) for the present suspension of polydisperse spherical particles. By contrast, the results \((3.56)_1\) and \((3.56)_4\) for the effective shear modulus \(\tilde{\mu}\) and effective electrostrictive coefficient \(\tilde{m}_K\) differ in general from expressions \((4.23)_1\) and \((4.23)_4\).

In the dilute limit of particles as \(c \to 0^+\), these latter expressions reduce asymptotically to

\[
\tilde{\mu}_{\text{DCS}} = \mu + \frac{5(3\kappa + 4\mu)(\mu_p - \mu)\mu}{(9\kappa + 8\mu)\mu + 6(\kappa + 2\mu)\mu_p} c,
\]

\[
\tilde{m}_{\text{DCS}} = \varepsilon + \frac{3(\varepsilon_p - \varepsilon)[6\mu_p(2\varepsilon + \varepsilon_p)(\kappa + 2\mu) + \mu(27\varepsilon\kappa + 28\varepsilon\mu - 4\varepsilon_p\mu)]\varepsilon}{(2\varepsilon + \varepsilon_p)^2[6\mu_p(\kappa + 2\mu) + \mu(9\kappa + 8\mu)]} c
\]

up to \(O(c)\). Thus, as anticipated in remark \(iv\) of Section 3.3, the result \((3.56)_1\) does agree identically with the exact effective shear modulus \((4.26)_1\) for a dilute suspension of spherical particles (cf. expression \((3.60)_1\)), but the same is not true for the result \((3.56)_4\), whose asymptotic form \((3.60)_4\) in the dilute limit is in general different from the effective electrostrictive coefficient \((4.26)_2\).

vii. While exact for a suspension with spherical particles of infinitely many sizes, the results \((4.23)_1\), \((4.23)_2\), \((4.23)_3\) have been shown (Segurado and LLorca, 2002; Lopez-Pamies et al., 2013b) to accurately describe as well the elastic and the dielectric response of isotropic suspensions with spherical particles of the same size up to relatively large particle concentrations sufficiently away from percolation (at least up to about \(c = 0.25\), but possibly up to larger values of \(c\) depending on the heterogeneity contrast). By means of sample comparisons with finite-element simulations generated with help of the framework presented in Chapter 5, we show in Chapter 6 that the results \((4.23)_4\) and \((4.23)_5\) for the electrostrictive coefficients \(\tilde{m}_K\) and \(\tilde{m}_J\) are also accurately descriptive of isotropic suspensions with spherical particles of the same size sufficiently away from percolation (at least up to about \(c = 0.2\)).
4.2 The overall electromechanical response of a differential coated cylinder assemblage

In this section, we derive the analytical solution for the electromechanical response of a *differential coated cylinder assemblage* (DCC) in the limit of small deformations and moderate electric fields. Again, we only consider here such transversely isotropic suspensions of aligned fibers with circular cross section wherein the matrix and the fillers are isotropic elastic dielectric materials. Based on practical grounds and to ease analytical tractability, we further restrict attention to material constituents that are ideal dielectrics (or equivalently, that do not exhibit “material” electrostriction, see remark *iii* above in Section 4.1). It follows that the tensors $L^{(r)}$, $\epsilon^{(r)}$, $M^{(r)}$ defined originally in (2.31) are now written as

$$L^{(1)} = 2\mu K + (3\lambda + 2\mu)J, \quad \epsilon^{(1)} = \varepsilon I, \quad M^{(1)} = \varepsilon K - \frac{\varepsilon}{2} J,$$

$$L^{(2)} = 2\mu_p K + (3\lambda_p + 2\mu_p)J, \quad \epsilon^{(2)} = \varepsilon_p I, \quad M^{(2)} = \varepsilon_p K - \frac{\varepsilon_p}{2} J. \quad (4.27)$$

In these expressions, we recall that $\mu, \mu_p, \lambda, \lambda_p, \varepsilon, \varepsilon_p$ stand for, respectively, the Lamé elastic moduli, permittivity, and electrostrictive coefficients of the matrix and the particles, and $K, J$ are two orthogonal projection tensors given by (3.20). Moreover, because of the assumed overall (geometric and constitutive) transverse isotropy of the suspension, we recall that the effective tensors (2.33)–(2.35) are of the form (3.39), hence fully defined by the thirteen effective constants $\tilde{c}_L, \tilde{d}_L, \tilde{e}_L, \tilde{f}_L, \tilde{g}_L, \tilde{\epsilon}_I, \tilde{\epsilon}_l, \tilde{c}_M, \tilde{d}_M, \tilde{e}_M, \tilde{f}_M, \tilde{g}_M, \tilde{h}_M$.

A coated cylinder assemblage is a two-phase particulate microstructure wherein aligned homothetic coated cylinders — comprising a cylindrical core of circular cross section made up of the fiber material that is surrounded by a cylindrical shell made up of the matrix material — of infinitely many sizes are assembled together to fill the entire domain $\Omega$ occupied by the composite (Hashin and Rosen, 1964). The particular manner in which the coated cylinders are assembled is arbitrary. Assemblages where coated cylinders of comparable radius are placed far apart from each other and surrounded by coated cylinders of much smaller radius, in such a way that the microstructure is fractal-like comprising a hierarchy of well-separated coated cylinders, are referred to as *differential coated cylinder* assemblages (Avellaneda, 1987).

One of two defining features of a differential coated cylinder assemblage is that, by construction, any coated cylinder in the assemblage can be regarded to be surrounded by a homogeneous medium
of infinite extent with the effective properties of the entire assemblage. The other defining feature being that the average response of any coated cylinder is the same as the average response of the entire assemblage (or, equivalently, that the so-called average “polarizability” of each coated cylinder vanishes); see Milton (1985) and Section 10.5 in Milton, 2002. These two defining features of such microstructures entail that the gradients of the tensor fields $\Gamma$ and $\gamma$ needed in expressions (2.33)–(2.35) to compute the effective tensors $\tilde{L}$, $\tilde{\epsilon}$, $\tilde{M}$ are the same in each of the coated cylinders. This allows to simplify expressions (2.33)–(2.35) to

$$
\tilde{L}_{ijkl} = \frac{1}{|C|} \int_{C} L_{ijpq} \Gamma_{pkl,q} dX,
$$

$$
\tilde{\epsilon}_{ij} = \frac{1}{|C|} \int_{C} \epsilon_{iq} \gamma_{q,j} dX,
$$

$$
\tilde{M}_{ijkl} = \frac{1}{|C|} \int_{C} \Gamma_{pij,q} M_{pqrs} \gamma_{r,k} \gamma_{s,l} dX,
$$

(4.28)

where the integrals are now over the domain $C$ occupied by a single coated cylinder, as opposed to over the entire domain $\Omega$. Moreover, these gradients can be expediently computed by taking the domain $\Omega$ in their defining boundary-value problems (2.36), (2.37) to be an infinite body comprised of a single coated cylinder, occupying the domain $C = \{X : |X - (X \cdot N)N| \leq 1\}$ say, embedded in a homogeneous medium with the effective properties of the entire assemblage. More specifically, when solving the boundary-value problem (2.36)–(2.37), it suffices to consider $\Omega = \mathbb{R}^3$, the local modulus of elasticity

$$
L = \begin{cases} 
L^{(2)} & \text{if } |X - (X \cdot N)N| \leq R_p \\
L^{(1)} & \text{if } R_p < |X - (X \cdot N)N| \leq 1 \\
\tilde{L} & \text{if } |X - (X \cdot N)N| > 1
\end{cases}
$$

(4.29)

the local permittivity tensor

$$
\epsilon = \begin{cases} 
\epsilon^{(2)} & \text{if } |X - (X \cdot N)N| \leq R_p \\
\epsilon^{(1)} & \text{if } R_p < |X - (X \cdot N)N| \leq 1 \\
\tilde{\epsilon} & \text{if } |X - (X \cdot N)N| > 1
\end{cases}
$$

(4.30)

where $R_p$ stands for the radius of the fiber within a coated cylinder of radius 1. We note for later reference that the local electrostrictive tensor is given by

$$
M = \begin{cases} 
M^{(2)} & \text{if } |X - (X \cdot N)N| \leq R_p \\
M^{(1)} & \text{if } R_p < |X - (X \cdot N)N| \leq 1 \\
\tilde{M} & \text{if } |X - (X \cdot N)N| > 1
\end{cases}
$$

(4.31)
For the class of dielectric elastomer composites of interest in this section, wherein the matrix and the fibers are isotropic ideal elastic dielectrics, the moduli of elasticity \( L(r) \), permittivity tensors \( \epsilon(r) \), and electrostrictive tensors \( M(r) \) \((x = 1, 2)\) are given by expressions (4.27).

Given the local modulus of elasticity and permittivity tensors (4.29)–(4.30), the solutions to the pdes (2.36) and (2.37) can be worked out in terms of solid harmonics (see, e.g., Bland, 1965). To simplify the calculations involved, it proves helpful to choose the direction of the fibers \( N \) to coincide with one of the laboratory axis. For \( N = e_3 \) say, the thirteen effective electromechanical constants (3.39) specialize to

\[
\begin{align*}
\bar{c}_L &= \bar{L}_{1111} + \bar{L}_{1122} \\
&= \frac{1}{|C|} \int_C L_{11pq} [\Gamma_{p11,q} + \Gamma_{p22,q}] \, dX, \\
\bar{d}_L &= \bar{L}_{3333} = \frac{1}{|C|} \int_C L_{33pq} \Gamma_{p33,q} \, dX, \\
\bar{e}_L &= 2\bar{L}_{1212} = \frac{2}{|C|} \int_C L_{12pq} \Gamma_{p12,q} \, dX, \\
\bar{f}_L &= 2\bar{L}_{1313} = \frac{2}{|C|} \int_C L_{13pq} \Gamma_{p13,q} \, dX, \\
\bar{g}_L &= \bar{L}_{1133} = \frac{1}{|C|} \int_C L_{11pq} \Gamma_{p33,q} \, dX; \\
\bar{\varepsilon}_t &= \bar{\varepsilon}_{11} = \frac{1}{|C|} \int_C \varepsilon_{\gamma 11,1} \, dX, \\
\bar{\varepsilon}_l &= \bar{\varepsilon}_{33} = \frac{1}{|C|} \int_C \varepsilon_{\gamma 33,3} \, dX; \\
\bar{c}_M &= \bar{M}_{1111} + \bar{M}_{2211} \\
&= \frac{1}{|C|} \int_C [\Gamma_{p11,q} + \Gamma_{p22,q}] M_{pqrs} \gamma_{r,1} \gamma_{s,1} \, dX, \\
\bar{d}_M &= \bar{M}_{3333} = \frac{1}{|C|} \int_C \Gamma_{p33,q} M_{pqrs} \gamma_{r,3} \gamma_{s,3} \, dX, \\
\bar{e}_M &= 2\bar{M}_{1212} = \frac{2}{|C|} \int_C \Gamma_{p12,q} M_{pqrs} \gamma_{r,1} \gamma_{s,2} \, dX, \\
\bar{f}_M &= 2\bar{M}_{1313} = \frac{2}{|C|} \int_C \Gamma_{p13,q} M_{pqrs} \gamma_{r,1} \gamma_{s,3} \, dX, \\
\bar{g}_M &= \bar{M}_{3311} = \frac{1}{|C|} \int_C \Gamma_{p33,q} M_{pqrs} \gamma_{r,1} \gamma_{s,1} \, dX, \\
\bar{h}_M &= \frac{1}{2} (\bar{M}_{1133} + \bar{M}_{2233}) \\
&= \frac{1}{|C|} \int_C [\Gamma_{p11,q} + \Gamma_{p22,q}] M_{pqrs} \gamma_{r,3} \gamma_{s,3} \, dX,
\end{align*}
\]

revealing that only certain combinations of the components of the gradients of the fields \( \Gamma \) and \( \gamma \)
are needed in the computation of these effective constants. In the sequel, for conciseness, we provide solutions only for these combinations.

### 4.2.1 The solution for \( \Gamma \)

We begin by presenting the solution for the components \( \Gamma_{112,j} \), which are needed in the computation of the effective elastic constant (4.32)\(_3\) and the effective electrostrictive constant (4.32)\(_{10}\). They read as

\[
\begin{align*}
\Gamma_{112,1} &= \left[ \frac{g'}{R} + 2h \right] X_1 X_2 + \frac{h'}{R} X_1^3 X_2, \\
\Gamma_{212,2} &= \left[ \frac{g'}{R} + 2h \right] X_1 X_2 + \frac{h'}{R} X_1 X_2^3, \\
\Gamma_{112,2} &= \Gamma_{212,1}, \\
\Gamma_{312,3} &= \Gamma_{312,1} = \Gamma_{212,3} = \Gamma_{312,2} = 0, \\
\end{align*}
\]

(4.33)

where \( g \) and \( h \) are functions of \( R = \sqrt{X_1^2 + X_2^2} \) given by

\[
g = \begin{cases} 
A_1 + A_3 R^2 & \text{if } 0 \leq R \leq R_p \\
B_1 + \frac{B_2}{R^4} + B_3 R^2 + \frac{B_4}{R^2} & \text{if } R_p < R \leq 1 
\end{cases}
\]

(4.34)

and

\[
h = \begin{cases} 
\frac{2(\lambda_p + 3\mu_p)}{2\lambda_p + 3\mu_p} A_3 & \text{if } 0 \leq R \leq R_p \\
-\frac{B_4}{R^8} - \frac{2(\lambda + 3\mu)}{2\lambda + 3\mu} B_3 + 2 \left[ 1 + \frac{\lambda}{\mu} \right] \frac{B_4}{R^3} & \text{if } R_p < R \leq 1 
\end{cases}
\]

(4.35)

In these expressions, use has been made of the notation \( g'(R) \equiv \frac{dg}{dR}, h'(R) \equiv \frac{dh}{dR} \), and \( A_1, A_3, B_1, B_2, B_3, B_4 \) are constants that depend on the volume fraction of fibers \( c = R_p^2 \), on the elastic properties of the matrix and the fibers, and on the effective elastic coefficients \( \tilde{c}_L \) and \( \tilde{e}_L \) of the entire assemblage. Because of their bulkiness, the explicit form of these constants is deferred to Appendix B.2.1.

We proceed with the solution for the combination of components \( \Gamma_{111,j} + \Gamma_{122,j} \). This combination is needed in the computation of the effective elastic constant (4.32)\(_1\), and the effective electrostrictive
constants \((4.32)_8\) and \((4.32)_{13}\). It can be written as follows:

\[
\begin{align*}
\Gamma_{111,1} + \Gamma_{122,1} &= \left[ d + \frac{e}{R^2} \right] - \frac{2e}{R^4} X_1^2, \\
\Gamma_{211,2} + \Gamma_{222,2} &= \left[ d + \frac{e}{R^2} \right] - \frac{2e}{R^4} X_2^2, \\
\Gamma_{111,2} + \Gamma_{122,2} &= \Gamma_{211,1} + \Gamma_{222,1} = -\frac{2e}{R^4} X_1 X_2, \\
\Gamma_{111,3} + \Gamma_{122,3} &= \Gamma_{311,1} + \Gamma_{322,1} = 0, \\
\Gamma_{211,3} + \Gamma_{222,3} &= \Gamma_{311,2} + \Gamma_{322,2} = 0, \\
\Gamma_{311,3} + \Gamma_{322,3} &= 0, \\
\end{align*}
\]

(4.36)

where \(d\) and \(e\) are given by

\[
d = \begin{cases} 
\frac{(\lambda + 2\mu)(\tilde{\epsilon}_L + \tilde{\delta}_L)}{\left(\lambda_p + \mu + \mu_p\right)[\tilde{\epsilon}_L + 2(\lambda + \mu)] - c(\lambda_p - \lambda + \mu_p - \mu)(\tilde{\delta}_L - 2\mu)} & \text{if } 0 \leq R \leq R_p \\
\frac{(\lambda + 2\mu)(\tilde{\epsilon}_L + \tilde{\delta}_L)}{\left(\lambda_p + \mu + \mu_p\right)[\tilde{\epsilon}_L + 2(\lambda + \mu)] - c(\lambda_p - \lambda + \mu_p - \mu)(\tilde{\delta}_L - 2\mu)} & \text{if } R_p < R \leq 1 
\end{cases}
\]

(4.37)

and

\[
e = \begin{cases} 
0 & \text{if } 0 \leq R \leq R_p \\
\frac{-c(\lambda_p - \lambda + \mu_p - \mu)(\tilde{\epsilon}_L + \tilde{\delta}_L)}{\left(\lambda_p + \mu + \mu_p\right)[\tilde{\epsilon}_L + 2(\lambda + \mu)] - c(\lambda_p - \lambda + \mu_p - \mu)(\tilde{\delta}_L - 2\mu)} & \text{if } R_p < R \leq 1 
\end{cases}
\]

(4.38)

Next, we record the solution for the components \(\Gamma_{i13,j}\) required in the computation of the effective elastic constant \((4.32)_4\) and the effective electrostrictive constant \((4.32)_{11}\). These components read as

\[
\begin{align*}
\Gamma_{113,3} &= \Gamma_{313,1} = t + \frac{u}{R^2} \left[ 1 - \frac{2X_1^2}{R^2} \right], \\
\Gamma_{213,3} &= \Gamma_{313,2} = -\frac{2uX_1X_2}{R^4}, \\
\Gamma_{113,1} &= \Gamma_{213,2} = \Gamma_{313,3} = \Gamma_{113,2} = \Gamma_{213,1} = 0, \\
\end{align*}
\]

(4.39)

where

\[
t = \begin{cases} 
\frac{2\mu \tilde{\epsilon}_L}{(\mu + \mu_p)(\tilde{\epsilon}_L + \tilde{\delta}_L) - c(\mu_p - \mu)(\tilde{\delta}_L - 2\mu)} & \text{if } 0 \leq R \leq R_p \\
\frac{\mu \tilde{\epsilon}_L}{(\mu + \mu_p)(\tilde{\epsilon}_L + \tilde{\delta}_L) - c(\mu_p - \mu)(\tilde{\delta}_L - 2\mu)} & \text{if } R_p < R \leq 1 
\end{cases}
\]

(4.40)

and

\[
u = \begin{cases} 
0 & \text{if } 0 \leq R \leq R_p \\
\frac{-c(\mu_p - \mu)\tilde{\delta}_L}{(\mu + \mu_p)(\tilde{\epsilon}_L + \tilde{\delta}_L) - c(\mu_p - \mu)(\tilde{\delta}_L - 2\mu)} & \text{if } R_p < R \leq 1 
\end{cases}
\]

(4.41)
Finally, we present the solution for the components $\Gamma_{i33,j}$, which are needed in the computation of the effective elastic constants $(4.32)_2$ and $(4.32)_5$, as well as of the effective electrostrictive constants $(4.32)_9$ and $(4.32)_{12}$. They read as

$$\Gamma_{133,1} = v + \frac{w}{R^2} \left[ 1 - 2 \frac{X_1^2}{R^2} \right],$$

$$\Gamma_{233,2} = v + \frac{w}{R^2} \left[ 1 - 2 \frac{X_2^2}{R^2} \right],$$

$$\Gamma_{133,2} = \Gamma_{233,1} = -2w \frac{X_1 X_2}{R^4},$$

$$\Gamma_{333,3} = 1,$$

$$\Gamma_{133,3} = \Gamma_{333,1} = \Gamma_{233,3} = \Gamma_{333,2} = 0,$$

where

$$v = \begin{cases} 
-2\frac{\lambda_0 + (1-c)\mu}{2(\lambda_0 + \mu + \mu_0)} (\mathbf{e}_L + 2(\lambda + \mu)) - 2c(\lambda_0 - \lambda + \mu + \mu_0) & \text{if } 0 \leq R \leq R_p \\
2(\lambda_0 + \mu + \mu_0)(\mathbf{g}_L - \lambda) + c(\lambda_0 - \lambda) & \text{if } R_p < R \leq 1 
\end{cases}$$

and

$$w = \begin{cases} 
0 & \text{if } 0 \leq R \leq R_p \\
-2c(\lambda_0 \mu - \lambda \mu_0 + (\lambda_0 - \lambda + \mu + \mu_0)\mathbf{e}_L) + c(\lambda_0 - \lambda) & \text{if } R_p < R \leq 1 
\end{cases}$$

### 4.2.2 The solution for $\gamma$

Having established the solution for the required components of the gradient of $\Gamma$, we now turn to present the solution for the components of the gradient of $\gamma$, which are needed in the computation of the two effective permittivity coefficients $(4.32)_6$ and $(4.32)_7$, as well as of the 6 effective electrostrictive coefficients $(4.32)_{8-13}$. They read as follows

$$\gamma_{1,1} = -\left[ a + \frac{b}{R^2} \right] + \frac{2b}{R^4} X_1^2,$$

$$\gamma_{2,2} = -\left[ a + \frac{b}{R^2} \right] + \frac{2b}{R^4} X_2^2,$$

$$\gamma_{1,2} = \gamma_{2,1} = \frac{2b}{R^4} X_1 X_2,$$

$$\gamma_{3,3} = 1,$$

$$\gamma_{1,3} = \gamma_{2,3} = \gamma_{3,1} = \gamma_{3,2} = 0,$$

(4.45)
where

\[ a = \begin{cases}
-4\varepsilon\varepsilon_t & \text{if } 0 \leq R \leq R_p \\
\frac{-4\varepsilon\varepsilon_t}{(\varepsilon_t + \varepsilon)(\varepsilon_p + \varepsilon) - c(\varepsilon_t - \varepsilon)(\varepsilon_p - \varepsilon)} & \text{if } R_p < R \leq 1
\end{cases} \tag{4.46} \]

and

\[ b = \begin{cases}
0 & \text{if } 0 \leq R \leq R_p \\
\frac{2c(\varepsilon_p - \varepsilon)\varepsilon_t}{(\varepsilon_t + \varepsilon)(\varepsilon_p + \varepsilon) - c(\varepsilon_t - \varepsilon)(\varepsilon_p - \varepsilon)} & \text{if } R_p < R \leq 1
\end{cases} \tag{4.47} \]

4.2.3 The effective electromechanical constants

We are now equipped to determine the thirteen effective constants that characterize the overall electromechanical response of the assemblage of coated cylinders. The integrals over a single coated cylinder in (4.32) can be readily carried out by making use of the explicit solutions (4.33), (4.36), (4.39), (4.42) for the combinations of components \( \Gamma_{i12,j}, \Gamma_{i11,j} + \Gamma_{i22,j}, \Gamma_{i13,j}, \Gamma_{i33,j} \) of the gradient of \( \Gamma \), as well as the components (4.45) of the gradient of \( \gamma \). Expressions (4.32)\_1–5 yield a system of coupled polynomial equations for the five effective elastic constants \( \tilde{c}_L, \tilde{a}_L, \tilde{e}_L, \tilde{f}_L, \tilde{g}_L \), from which a unique solution can be extracted based on physical arguments. Expression (4.32)\_6 also yields a polynomial equation for the effective permittivity coefficient \( \tilde{\epsilon}_t \) from which a unique solution can be deduced. On the other hand, expressions (4.32)\_7–13 render explicit solutions for the effective permittivity coefficient \( \tilde{\epsilon}_t \) and the effective electrostrictive constants \( \tilde{c}_M, \tilde{d}_M, \tilde{e}_M, \tilde{f}_M, \tilde{g}_M, \tilde{h}_M \).

After some algebraic manipulation, the result for all thirteen effective electromechanical constants
can be written as

\[\tilde{c}_{DCC}^{L} = 2\lambda + 2\mu + \frac{2c(\lambda + 2\mu)(\Delta \lambda + \Delta \mu)}{\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)},\]

\[\tilde{d}_{DCC}^{L} = \lambda + 2\mu + c(\Delta \lambda + 2\Delta \mu) - \frac{c(1 - c)\Delta \lambda^2}{\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)},\]

\[\tilde{c}_{L}^{DCC} = 2q_2 + \frac{\sqrt{q_2^2 + q_1 q_3}}{q_1} \mu, \quad \tilde{\varepsilon}_{DCC}^{L} = 2\mu + \frac{4c\mu\Delta \mu}{2\mu + (1 - c)\Delta \mu},\]

\[\tilde{g}_{L}^{DCC} = \lambda + \frac{c\Delta \lambda (\lambda + 2\mu)}{\lambda + 2\mu + (1 - c)(\Delta \lambda + \Delta \mu)}; \quad \tilde{\varepsilon}_{e}^{DCC} = \varepsilon + c\Delta \varepsilon; \quad \tilde{\varepsilon}_{l}^{DCC} = \varepsilon + c\Delta \varepsilon; \quad \tilde{\varepsilon}_{l}^{DCC} = \varepsilon + c\Delta \varepsilon; \quad \tilde{h}_{M}^{DCC} = 0,\]

where the superscript “DCC” has been appended for clarity. In these expressions, it is recalled that \(\Delta \lambda = \lambda_p - \lambda, \Delta \mu = \mu_p - \mu, \Delta \varepsilon = \varepsilon_p - \varepsilon\), the constants \(a_p, a_n, b_n\) are defined in expressions (4.46) and (4.47), the constants \(A_1, A_3, B_1, B_2, B_3, B_4\) are defined in Appendix B.2.1, and the final set of constants \(q_1, q_2, q_3\), which depend explicitly on the volume fraction of fibers \(c\) and the elastic properties of the matrix and the fibers, are given by expressions (B.5) in Appendix B.2.2. A few comments are in order:

1. With the exception of the effective elastic constant \(\tilde{c}_{L}^{DCC}\) and the effective electrostrictive constant \(\tilde{e}_{M}^{DCC}\), all of the remaining eleven effective electromechanical constants (4.48) agree identically with the result (3.66) for the iterative microstructure of Chapter 3. This agreement is admittedly remarkable since the two sets of results pertain to two different microstructures. These microstructures, however, do have the same one- and two-point correlations functions. Furthermore, these microstructures are similar in that the fibers in both of them can act as “neutral inclusions” under the same type of loading conditions (see Appendix B in Lopez-Pamies, 2014).
ii. We recall here that the five effective elastic constants $\tilde{c}_L^{DCC}, \tilde{d}_L^{DCC}, \tilde{e}_L^{DCC}, \tilde{f}_L^{DCC}, \tilde{g}_L^{DCC}$ can be recast into the five more conventional elastic parameters, $\tilde{\mu}_t^{DCC}, \tilde{\kappa}_t^{DCC}, \tilde{\mu}_l^{DCC}, \tilde{E}_l^{DCC}, \tilde{\nu}_l^{DCC}$, utilized to characterize the elastic response of transversely isotropic materials, via relations (3.41). In this connection, it is not difficult to verify that the effective transverse shear modulus $\tilde{\mu}_t^{DCC}$ agrees identically with the result originally derived by Christensen and Lo (1979), and later proved to be realizable by Avellaneda (1987), for a differential coated cylinder assemblage. Further, the remaining four effective elastic constants $\tilde{\kappa}_t^{DCC}, \tilde{\mu}_l^{DCC}, \tilde{E}_l^{DCC}, \tilde{\nu}_l^{DCC}$ agree identically with the classical results of Hashin and Rosen (1964) and Hill (1964) for arbitrary (not necessarily differential) coated cylinder assemblages. Similarly, the two effective permittivities $\tilde{\varepsilon}_t^{DCC}$ and $\tilde{\varepsilon}_l^{DCC}$ agree identically with the classical results (see Hashin (1979) and references therein) for arbitrary (not necessarily differential) coated cylinder assemblages.

iii. Via a neutral inclusion argument, the effective electrostrictive coefficients $\tilde{c}_M^{DCC}, \tilde{d}_M^{DCC}, \tilde{f}_M^{DCC}, \tilde{g}_M^{DCC}, \tilde{h}_M^{DCC}$ can be shown to be exact for arbitrary (not necessarily differential) coated cylinder assemblages. By contrast, this is not true for the effective electrostrictive coefficient $\tilde{e}_M^{DCC}$, which is only valid for differential coated cylinder assemblages.
Hybrid FE formulation for the effective response of dielectric elastomer composites in the small-deformation limit

If you are lucky enough to have lived in Paris as a young man, then wherever you go for the rest of your life, it stays with you, for Paris is a moveable feast.


Despite the discontinuous and oscillating character of their coefficients, the boundary value problems (2.36) and (2.37) for the tensors Γ and γ — needed, again, in the computation of the effective electromechanical tensors (2.33)–(2.35) — are elliptic. As such, they can be expediently solved numerically via the finite-element (FE) method. In this chapter, we present FE formulations to solve the pdes (2.36) and (2.37) and compute the effective electromechanical tensors $\tilde{L}$, $\tilde{\epsilon}$, and $\tilde{M}$. The hybrid nature of this framework that we adopt here allows for the analysis of compressible, as well as incompressible behaviors typical of elastomers. Complementary to the two previous chapters wherein the underlying filler particles are infinitely polydisperse in size, the FE framework is presented within the context of spherical particles of monodisperse size and aligned fibers of circular cross section and of monodisperse size.
5. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

5.1 The overall electromechanical response of a distribution of spherical particles

In this section, we provide details of the finite element framework employed to obtain the overall electromechanical response of dielectric elastomers filled with a random distribution of spherical particles of monodisperse size.

While the formulation (2.33)–(2.37) is formal in general, Tian (2007) and Tian et al. (2012) have shown it to be rigorous in the context of composite materials with periodic microstructure. When the repeating unit cell \( Q \) chosen to describe such a periodic microstructure is the unit cube \( Q = \{ X \in [0, 1]^3 \} \), the general formulation (2.33)–(2.37) specializes to

\[
\tilde{L}_{ijkl} = \int_Q L_{ijmn} \Gamma_{mkl,n} \, dX, \quad (5.1)
\]

\[
\tilde{\epsilon}_{ij} = \int_Q \epsilon_{im} \gamma_{m,j} \, dX, \quad (5.2)
\]

\[
\tilde{M}_{ijkl} = \int_Q \Gamma_{mij,n} M_{mnpq} \gamma_{p,k} \gamma_{q,l} \, dX, \quad (5.3)
\]

where now the tensor fields \( \Gamma \) and \( \gamma \) are defined by

\[
[L_{ijmn} \Gamma_{mkl,n}]_{,j} = 0, \quad X \in Q \quad \text{with} \quad \begin{cases} 
\Gamma_{ikl}(1, X_2, X_3) - \Gamma_{ikl}(0, X_2, X_3) = \delta_{ik} \delta_{l1} \\
\Gamma_{ikl}(X_1, 1, X_3) - \Gamma_{ikl}(X_1, 0, X_3) = \delta_{ik} \delta_{l2}, \quad X \in \partial Q \\
\Gamma_{ikl}(X_1, X_2, 1) - \Gamma_{ikl}(X_1, X_2, 0) = \delta_{ik} \delta_{l3}
\end{cases} \quad (5.4)
\]

and

\[
[\epsilon_{im} \gamma_{m,j}]_{,i} = 0, \quad X \in Q \quad \text{with} \quad \begin{cases} 
\gamma_i(1, X_2, X_3) - \gamma_i(0, X_2, X_3) = \delta_{i1} \\
\gamma_i(X_1, 1, X_3) - \gamma_i(X_1, 0, X_3) = \delta_{i2}, \quad X \in \partial Q \\
\gamma_i(X_1, X_2, 1) - \gamma_i(X_1, X_2, 0) = \delta_{i3}
\end{cases} \quad (5.5)
\]

with \( \partial Q \) denoting the boundary of \( Q \). It is plain that the partial differential equations (5.4)–(5.5), or more generally (2.36)–(2.37), do not admit explicit solutions other than for special cases (see, e.g., Section 4.1). However, being second-order linear elliptic pdes, they can be readily solved numerically by means of the finite element method, as outlined next.

FE formulation. One of the foci of this dissertation is on the practical case of dielectric elastomers filled with a random and isotropic distribution of spherical particles. To approximate the randomness
5. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

and isotropy of this class of microstructures, following common practice (see, e.g., Gusev, 1997; Michel et al., 1999), we begin by considering an infinite periodic medium where the repeated unit cell comprises a large but finite number \( N \) of randomly distributed spherical particles. Previous results based on this approach — for linear (Segurado and LLorca, 2002) as well as for nonlinear (Lopez-Pamies et al., 2013b) problems — have indicated that \( N = 30 \) particles is, in general, sufficient to approximate the isotropy of the microstructure. Figure 5.1 depicts representative examples of such unit cells with \( N = 30 \) particles for various particle concentrations: (a) \( c = 0.05 \), (b) \( c = 0.15 \), and (c) \( c = 0.25 \). Details on the adsorption algorithm employed to generate such microstructures are deferred to Section 7.2.2. Having identified the microstructures of interest, we carry out their discretizations by means of the mesh generator code Netgen (Schöberl, 1997). Hybrid isoparametric 10-node tetrahedral elements with constant pressure were selected to solve the pde (5.4), while isoparametric 10-node tetrahedral elements were utilized for the pde (5.5). To avoid loss of continuity, the convergence properties of this choice of FE discretization are deferred to Appendix C.1. Figure 5.2 displays three meshes of increasing refinement with particle concentration \( c = 0.15 \). Mesh sensitivity analyses revealed that meshes containing about 400,000 elements (\( \sim 570,000 \) nodes), such as the one shown in Fig. 5.2(c), are refined enough to deliver accurate results.

The resulting discretized version of equations (5.4)–(5.5) for \( \Gamma \) and \( \gamma \) are formulated and solved via a vectorized FE code written in the technical computing environment MATLAB (see MATLAB Version 8.3 Documentation, 2014). The computed FE solutions for the fields \( \Gamma \) and \( \gamma \) are then utilized in the definitions (5.1)–(5.3) to finally compute the three effective tensors that describe the macroscopic electromechanical response of the pertinent dielectric elastomer composite: \( \mathbf{\tilde{L}}^{FE}, \mathbf{\varepsilon}^{FE}, \)
and $\tilde{M}^{FE}$ (here and subsequently, for clarity, the superscript “FE” is appended to any quantity based on FE solutions).

**Assessment of the simulated microstructures.** Because of the finite number of particles ($N = 30$) included per unit cell, the microstructures simulated here are (not exactly but) only approximately isotropic. In order to assess the isotropy of each realization that is constructed, we monitor the following measures

\[
\begin{align*}
    d_L &= \frac{\|2\tilde{\mu}^{FE} K + 3\tilde{\kappa}^{FE} J - \tilde{L}^{FE}\|_\infty}{\|\tilde{L}^{FE}\|_\infty}, \\
    d_\varepsilon &= \frac{\|\tilde{\varepsilon}^{FE} I - \tilde{\varepsilon}^{FE}\|_\infty}{\|\tilde{\varepsilon}^{FE}\|_\infty}, \\
    d_M &= \frac{\|\tilde{m}_K^{FE} K + \tilde{m}_J^{FE} J - \tilde{M}^{FE}\|_\infty}{\|\tilde{M}^{FE}\|_\infty},
\end{align*}
\]

where the effective electromechanical coefficients $\tilde{\mu}^{FE}$, $\tilde{\kappa}^{FE}$, $\tilde{\varepsilon}^{FE}$, $\tilde{m}_K^{FE}$, and $\tilde{m}_J^{FE}$ are defined by

\[
\begin{align*}
    \tilde{\mu}^{FE} &= \frac{1}{10} K \cdot \tilde{L}^{FE} = \frac{1}{20} \left( \tilde{L}^{FE}_{ijij} + \tilde{L}^{FE}_{ijji} - 2 / 3 \tilde{L}^{FE}_{iijj} \right), \\
    \tilde{\kappa}^{FE} &= \frac{1}{3} J \cdot \tilde{L}^{FE} = \frac{1}{9} \tilde{L}^{FE}_{iijj}, \\
    \tilde{\varepsilon}^{FE} &= \frac{1}{3} I \cdot \tilde{\varepsilon}^{FE} = \frac{1}{3} \tilde{\varepsilon}^{FE}, \\
    \tilde{m}_K^{FE} &= \frac{1}{5} K \cdot \tilde{M}^{FE} = \frac{1}{10} \left( \tilde{M}^{FE}_{ijij} + \tilde{M}^{FE}_{ijji} - 2 / 3 \tilde{M}^{FE}_{iijj} \right), \\
    \tilde{m}_J^{FE} &= J \cdot \tilde{M}^{FE} = \frac{1}{3} \tilde{M}^{FE}_{iijj},
\end{align*}
\]

and it is recalled that $K$ and $J$ stand for the orthogonal projection tensors given by expressions (3.20). For the class of dielectric elastomer composites of interest in Section 5, wherein the matrix and particles are both isotropic elastic dielectrics, a perfectly isotropic microstructure would result.
5. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

In measures \( d_L = d_\varepsilon = d_M = 0 \). In this dissertation, only realizations with

\[
\max\{d_L, d_\varepsilon, d_M\} \leq 10^{-2}
\]

are admitted as approximately isotropic.

5.2 The overall electromechanical response of a distribution of aligned fibers

In this section, we provide details of the FE framework employed to obtain the overall electromechanical response of dielectric elastomers filled with a random distribution of aligned fibers with monodisperse circular cross section.

We recall that the general formulae (2.33)–(2.35) for the effective electromechanical tensors \( \tilde{L}, \tilde{\varepsilon}, \tilde{M} \) of dielectric elastomer composites with arbitrary heterogeneous local modulus of elasticity, \( L = L(X) \), permittivity, \( \varepsilon = \varepsilon(X) \), and electrostrictive tensor, \( M = M(X) \), require solving the three-dimensional boundary-value problems (2.36)–(2.37) for the tensor fields \( \Gamma \) and \( \gamma \). Now, for microstructures comprising aligned cylindrical fibers, the heterogeneity is two-dimensional. By choosing — as in the foregoing and without loss of generality — the direction of the fibers \( N \) to coincide with the laboratory axis \( e_3 \) (see Fig. 6.7), the local electromechanical tensors for the problem of interest here read as

\[
\begin{align*}
L(X) &= [3\lambda(X_1, X_2) + 2\mu(X_1, X_2)]I + 2\mu(X_1, X_2)K, \\
\varepsilon(X) &= \varepsilon(X_1, X_2)I, \\
M(X) &= \varepsilon(X_1, X_2) \left( K - \frac{\mathcal{J}}{2} \right).
\end{align*}
\]

(5.13)

Because of the isotropy of these tensors and their independence of the \( X_3 \) coordinate, it follows that the three-dimensional boundary-value problems (2.36)–(2.37) can actually be recast as two-dimensional boundary-value problems:
The isotropy of the local modulus of elasticity (5.13) and its independence from $X_3$ imply that

\[
\begin{align*}
\Gamma_{\eta11,\iota} &= \Gamma_{\eta11,\iota}(X_1, X_2), \\
\Gamma_{\eta22,\iota} &= \Gamma_{\eta22,\iota}(X_1, X_2), \\
\Gamma_{\eta12,\iota} &= \Gamma_{\eta12,\iota}(X_1, X_2), \\
\Gamma_{\eta21,\iota} &= \Gamma_{\eta21,\iota}(X_1, X_2), \\
\Gamma_{\eta33,\iota} &= \Gamma_{\eta33,\iota}(X_1, X_2),
\end{align*}
\]

and that $\Gamma_{\eta11, \Gamma_{\eta22, \Gamma_{\eta12, \Gamma_{\eta21, \Gamma_{\eta33}}}}}$ are solutions to

\[
\begin{align*}
[\lambda(X_1, X_2)\Gamma_{\eta kl, \iota} + 2\mu(X_1, X_2)\Gamma_{\eta kl, \iota}]_{,\iota} &= 0, \quad X \in \Omega, \\
\text{with } \Gamma_{\eta kl} &= \delta_{\eta k}X_l, \quad X \in \partial\Omega,
\end{align*}
\]

where the pair of indices $(kl)$ are to be evaluated as 11, 22, 12, 21, 33. They further imply that

\[
\begin{align*}
\Gamma_{3\eta3,\iota} &= \Gamma_{3\eta3,\iota}(X_1, X_2), \\
\Gamma_{33\eta,\iota} &= \Gamma_{33\eta,\iota}(X_1, X_2), \\
\Gamma_{\eta13,\iota} &= \Gamma_{313,\iota} = \Gamma_{\eta31,\iota} = \Gamma_{331,\iota} = 0, \\
\Gamma_{\eta23,\iota} &= \Gamma_{323,\iota} = \Gamma_{\eta32,\iota} = \Gamma_{332,\iota} = 0,
\end{align*}
\]

and $\Gamma_{3\eta3}, \Gamma_{33\eta}$ are solutions to

\[
\begin{align*}
\begin{align*}
[\mu(X_1, X_2)\Gamma_{3 kl, \iota}]_{,\iota} &= 0, \quad X \in \Omega, \\
\text{with } \Gamma_{3 kl} &= \delta_{3 k}X_l, \quad X \in \partial\Omega,
\end{align*}
\end{align*}
\]

where now the pair of indices $(kl)$ are to be evaluated as 13, 31, 23, 31. In expressions (5.14)–(5.17) and subsequently, Greek indices range from 1 to 2.

Similarly, the isotropy of the local permittivity (5.13) and its independence from $X_3$ imply that

\[
\begin{align*}
\gamma_{\eta,\iota} &= \gamma_{\eta,\iota}(X_1, X_2), \\
\gamma_{\eta,\iota} &= \gamma_{r,\iota} = 0, \quad \gamma_{3,3} = 1,
\end{align*}
\]

and $\gamma_\eta$ are solutions to

\[
\begin{align*}
\begin{align*}
[\varepsilon(X_1, X_2)\gamma_{\eta,\iota}]_{,\eta} &= 0, \quad X \in \Omega, \quad \text{with } \gamma_{\eta} = X_\eta, \quad X \in \partial\Omega.
\end{align*}
\end{align*}
\]
It is plain that the boundary-value problems (5.15), (5.17), (5.19), do not admit explicit solutions other than for special cases (see, e.g., Section 4.2). However, the underlying pdes being second-order linear elliptic, it is straightforward to solve them numerically by means of the finite element method, as outlined next.

**FE formulation.** Similar to Section 3.6, we focus here on dielectric elastomers strengthened/weakened by transversely isotropic distributions of aligned cylindrical fibers with circular cross section. To approximate the transverse isotropic of this class of microstructures, and in view of the two dimensionality of the boundary-value problems (5.15), (5.17), (5.19), we consider an infinite two-dimensional periodic medium described by the periodic repetition of a square unit cell, $S = \{ \mathbf{X} : X_i \in [0, 1] \ i = 1, 2 \}$, that comprises a large but finite number $N$ of randomly distributed circles. Previous results based on this approach (see, e.g., Moraleda et al., 2009) have indicated that $N = 60$ circles is, in general, sufficient to approximate the transverse isotropy of the microstructure. Figure 5.3 depicts representative examples of such unit cells with $N = 60$ circles for various volume fractions of fibers: (a) $c = 0.05$, (b) $c = 0.15$, and (c) $c = 0.25$. Details on the adsorption algorithm employed to generate such microstructures are not reported here but can be deduced from its three-dimensional counterpart employed in Section 5.1 and described in Section 7.2.2. For the above-described repeating unit cells $S$, the boundary value problems (5.15), (5.17),

Figure 5.3: Unit cells with $N = 60$ circles randomly distributed for three volume fractions of fibers: (a) $c = 0.05$, (b) $c = 0.15$, and (c) $c = 0.25$. 
5. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

Figure 5.4: Three progressively refined meshes, with approximately (a) 8,000, (b) 40,000, and (c) 200,000 elements, of a unit cell containing a random distribution of \( N = 60 \) circles at volume fraction \( c = 0.15 \).

(5.19) specialize to

\[
\lambda(X_1, X_2) \Gamma_{\eta k l, \nu} \delta_{\eta \nu} + 2\mu(X_1, X_2) \Gamma_{\eta k l, \nu}, \delta_{\eta \nu} = 0, \quad \forall X \in S, \tag{5.20}
\]

with

\[
\begin{align*}
\Gamma_{\eta k l}(1, X_2) - \Gamma_{\eta k l}(0, X_2) &= \delta_{\eta k} \delta_{l 1} \\
\Gamma_{\eta k l}(X_1, 1) - \Gamma_{\eta k l}(X_1, 0) &= \delta_{\eta k} \delta_{l 2}
\end{align*}
, \quad \forall X \in \partial S,
\]

\[
\mu(X_1, X_2) \Gamma_{3 k l, \nu}, \delta_{\eta \nu} = 0, \quad \forall X \in S, \tag{5.21}
\]

with

\[
\begin{align*}
\Gamma_{3 k l}(1, X_2) - \Gamma_{3 k l}(0, X_2) &= \delta_{3 k} \delta_{l 1} \\
\Gamma_{3 k l}(X_1, 1) - \Gamma_{3 k l}(X_1, 0) &= \delta_{3 k} \delta_{l 2}
\end{align*}
, \quad \forall X \in \partial S,
\]

and

\[
\varepsilon(X_1, X_2) \gamma_{\eta, \nu}, \delta_{\eta \nu} = 0, \quad \forall X \in S, \tag{5.22}
\]

with

\[
\begin{align*}
\gamma_{\eta}(1, X_2) - \gamma_{\eta}(0, X_2) &= \delta_{\eta 1} \\
\gamma_{\eta}(X_1, 1) - \gamma_{\eta}(X_1, 0) &= \delta_{\eta 2}
\end{align*}
, \quad \forall X \in \partial S.
\]

In the above expressions, \( \partial S \) stands for the boundary of \( S \) and it is recalled that the pair of indices \( (kl) \) are to be evaluated as \( 11, 22, 12, 21, 33 \) in (5.20) and as \( 13, 31, 23, 31 \) in (5.21).

We carry out the discretization of these problems with help of the scripting and meshing capabilities of the FE package ABAQUS (see, ABAQUS Version 6.11 Documentation, 2011). Hybrid isoparametric 8-node quadrilateral elements with linearly varying pressure were selected to solve the pde (5.20), while isoparametric 8-node quadrilateral elements were utilized for the pdes (5.21).
and (5.22). Again, to avoid loss of continuity, the convergence properties of this choice of FE discretization are reported in Appendix C.2. Figure 5.4 display three meshes of increasing refinement for volume fraction of fibers $c = 0.15$. Mesh sensitivity analyses revealed that meshes containing about 200,000 elements ($\sim 600,000$ nodes), such as the one shown in Fig. 5.4(c), are refined enough to deliver accurate results. The discretized equations resulting from (5.20), (5.21), (5.22) are formulated and solved via a vectorized FE code written in the technical computing environment MATLAB (see MATLAB Version 8.3 Documentation, 2014). The computed FE solutions for the components of the tensor fields $\mathbf{\Gamma}$ and $\mathbf{\gamma}$ are then utilized to finally compute the three sought after effective electromechanical tensors via

\[
\tilde{L}_{ijkl}^{FE} = \int_S L_{ijpq}^{FE} \Gamma_{pklq}^{FE} \, d\mathbf{X},
\]
\[
\tilde{\epsilon}_{ij}^{FE} = \int_S \epsilon_{iq}^{FE} \gamma_{ jq}^{FE} \, d\mathbf{X},
\]
\[
\tilde{M}_{ijkl}^{FE} = \int_S \Gamma_{pjiq}^{FE} M_{pqvw}^{FE} \gamma_{vk}^{FE} \gamma_{w}^{FE} \, d\mathbf{X},
\]

where the superscript “FE” has been appended to these expressions for clarity.

**Assessment of the simulated microstructures.** Because of the finite number of fibers ($N = 60$) included per unit cell, the microstructures simulated here are transversely isotropic only in an approximate sense. In order to assess the transverse isotropy of each realization that is constructed, we monitor the following measures

\[
\delta_{\mathbf{L}} = \frac{\|\tilde{c}_{\mathbf{L}}^{FE} \mathbf{\epsilon}^{(1)} + \tilde{d}_{\mathbf{L}}^{FE} \mathbf{\epsilon}^{(2)} + \tilde{e}_{\mathbf{L}}^{FE} \mathbf{\epsilon}^{(3)} + \tilde{f}_{\mathbf{L}}^{FE} \mathbf{\epsilon}^{(4)} + \tilde{g}_{\mathbf{L}}^{FE} (\mathbf{\epsilon}^{(5)} + \mathbf{\epsilon}^{(6)}) - \tilde{\mathbf{L}}^{FE} \|_\infty}{\|\mathbf{L}^{FE} \|_\infty},
\]
\[
\delta_{\mathbf{\epsilon}} = \frac{\|\tilde{\epsilon}_{\mathbf{H}}^{FE} (\mathbf{I} - \mathbf{N} \otimes \mathbf{N}) + \tilde{f}_{\mathbf{H}}^{FE} \mathbf{N} \otimes \mathbf{N} - \tilde{\mathbf{\epsilon}}^{FE} \|_\infty}{\|\tilde{\mathbf{\epsilon}}^{FE} \|_\infty},
\]
\[
\delta_{\mathbf{M}} = \frac{\|\tilde{c}_{\mathbf{M}}^{FE} \mathbf{\epsilon}^{(1)} + \tilde{d}_{\mathbf{M}}^{FE} \mathbf{\epsilon}^{(2)} + \tilde{e}_{\mathbf{M}}^{FE} \mathbf{\epsilon}^{(3)} + \tilde{f}_{\mathbf{M}}^{FE} \mathbf{\epsilon}^{(4)} + \tilde{g}_{\mathbf{M}}^{FE} \mathbf{\epsilon}^{(5)} + \tilde{h}_{\mathbf{M}}^{FE} \mathbf{\epsilon}^{(6)} - \tilde{\mathbf{M}}^{FE} \|_\infty}{\|\mathbf{M}^{FE} \|_\infty},
\]

where the effective electromechanical coefficients $\tilde{c}_{\mathbf{L}}^{FE}, \tilde{d}_{\mathbf{L}}^{FE}, \tilde{e}_{\mathbf{L}}^{FE}, \tilde{f}_{\mathbf{L}}^{FE}, \tilde{g}_{\mathbf{L}}^{FE}, \tilde{\epsilon}_{\mathbf{L}}^{FE}, \tilde{\epsilon}_{\mathbf{H}}^{FE}, \tilde{c}_{\mathbf{M}}^{FE}, \tilde{d}_{\mathbf{M}}^{FE}$, and $\tilde{\mathbf{M}}^{FE}$, and $\tilde{\mathbf{L}}^{FE}$ are computed from the discretized equations.
Here, we remark the fourth order tensors $\mathbf{\epsilon}^{(i)}$ defined in (3.40) are to be evaluated with $\mathbf{N} = e_3$. For the class of dielectric elastomer composites of interest in Section 4, wherein the matrix and particles are both isotropic elastic dielectrics, a perfectly transversely isotropic microstructure would result in measures $\delta_L = \delta_\epsilon = \delta_M = 0$. Here, only realizations with

$$\max\{\delta_L, \delta_\epsilon, \delta_M\} \leq 10^{-2}$$

are admitted as approximately transversely isotropic.
Sample results in the limit of small deformations and moderate electric fields

The first principle is that you must not fool yourself and you are the easiest person to fool.

– Richard P. Feynman, Surely You’re Joking, Mr. Feynman!, 1984

To shed light on recent experimental findings, we deploy in this chapter the analytical and numerical results derived in Chapters 3, 4, and 5 to examine the elastic dielectric response of dielectric elastomers filled with isotropic distributions of stiff high-permittivity spherical particles. With the aim of identifying what other type of fillers not yet utilized in experiments may potentially lead to enhanced behaviors, we also present corresponding sample results for high-permittivity filler particles that are liquid-like in mechanical behavior and for the case when the fillers are vacuous pores. With the objective of also gaining insight into the effect that the addition of anisotropic fillers can have on the electromechanical properties of elastomers, analogous sample results are presented for the case of elastomers filled with aligned cylindrical fibers.

6.1 Isotropic dielectric elastomers filled with spherical particles

We recall that the practical motivation to consider isotropic distributions of spherical particles stems from recent experimental findings, including those of Zhang et al. (2002), Huang and Zhang (2004), Huang et al. (2005), Carpi and De Rossi (2005), Mc Carthy et al. (2009), and Liu et al. (2013), which have shown that the addition of random distributions of roughly spherical particles, made
up of high-permittivity or (semi-)conducting solids, into dielectric elastomers leads to a drastic
enhancement of the electrostrictive capabilities of these materials. Furthermore, we seek to identify
what other type of fillers not yet utilized in experimental studies, such as liquid-like particles with
high-permittivity and vacuous pores, may potentially lead to the enhancement of the overall elastic
dielectric properties of dielectric elastomers.

6.1.1 Electrostriction

In the absence of applied stresses when \( \overline{S} = 0 \), it follows from the overall constitutive relation (3.58)
for isotropic dielectric elastomer composites that

\[
\mathbf{H} = \frac{\tilde{m}_K}{2\mu} \mathbf{E} \otimes \mathbf{E} + \left[ \frac{\tilde{m}_K}{6\mu} - \frac{\tilde{m}_J}{9\kappa} \right] (\mathbf{E} \cdot \mathbf{E}) \mathbf{I}.
\]

The deformation measure (6.1) is referred to as the electrostriction that the dielectric elastomer
composite undergoes when it is subjected to a macroscopic electric field \( \overline{E} \).

In practice, it is often the case that just a uniaxial electric field is applied to probe the elec-
trostriction of deformable dielectrics. This is commonly accomplished by sandwiching a thin layer
of the material in between two compliant electrodes connected to a battery. For such a configura-
tion, the macroscopic stress is indeed roughly zero everywhere (inside the material as well as in the
surrounding space), while the macroscopic electric field is roughly uniform within the material and
zero outside of it. For an applied uniaxial electric field of the form

\[
\overline{E} = \bar{E} \mathbf{e}_3
\]

with \( \bar{E} = \Phi / L_3 \), where \( \Phi \) denotes the voltage applied between the electrodes and \( L_3 \) stands for the
initial thickness of the thin layer of dielectric elastomer composite, the electrostriction (6.1) takes
the diagonal form

\[
\mathbf{H} = \mathcal{H}_{11} \mathbf{e}_1 \otimes \mathbf{e}_1 + \mathcal{H}_{22} \mathbf{e}_2 \otimes \mathbf{e}_2 + \mathcal{H}_{33} \mathbf{e}_3 \otimes \mathbf{e}_3,
\]

where

\[
\mathcal{H}_{11} = \mathcal{H}_{22} = \left( \frac{\tilde{m}_K}{6\mu} - \frac{\tilde{m}_J}{9\kappa} \right) \bar{E}^2 \quad \text{and} \quad \mathcal{H}_{33} = - \left( \frac{\tilde{m}_K}{3\mu} + \frac{\tilde{m}_J}{9\kappa} \right) \bar{E}^2.
\]

Figure 6.1 shows a schematic of this thought experiment with the various quantities of interest
indicated.
For later reference, we note that in the absence of particles \( c = 0 \) expressions (6.4) reduce to
\[
H_{m11}^p = H_{m22}^p = \left( \frac{\varepsilon_6}{6\mu} + \frac{\varepsilon_18}{18\kappa} \right) E^2
\]
and
\[
H_{m33}^p = -\left( \frac{\varepsilon_3}{3\mu} - \frac{\varepsilon_18}{18\kappa} \right) E^2,
\]
the electrostriction in the transverse and thickness directions of the pure (unfilled) elastomeric matrix. The ratios
\[
\frac{H_{11}}{H_{11}^p} = \frac{H_{22}}{H_{22}^p} = \frac{\tilde{m}_K}{2\mu} - \frac{\tilde{m}_J}{3\kappa}, \quad \text{and} \quad \frac{H_{33}}{H_{33}^p} = \frac{\tilde{m}_K}{\mu} + \frac{\tilde{m}_J}{\kappa}
\]
provide then direct insight into how the addition of particles affects the electrostriction of dielectric elastomers.

### 6.1.2 Results for stiff particles with high permittivity

We begin by examining the case of dielectric elastomer composites wherein the filler particles are mechanically stiffer than the elastomeric matrix and also exhibit higher permittivity. As pointed out above, this is the case that has hitherto received most attention by the experimental community, presumably because most filler materials with high permittivity or (semi-)conducting behavior (e.g., ceramics, metals) are stiffer than elastomers.

Figures 6.2(a), (b), (c) show results for the normalized effective shear modulus \( \tilde{\mu}/\mu \), permittivity \( \tilde{\varepsilon}/\varepsilon \), and electrostrictive coefficients \( \tilde{m}_K/\varepsilon \), \( -2\tilde{m}_J/\varepsilon \) of a dielectric elastomer composite comprised of a nearly incompressible elastomeric matrix with \( \kappa/\mu = 10^3 \) and stiff high-permittivity filler particles with \( \mu_p/\mu = 10^5 \), \( \kappa_p/\mu = 10^5 \), and \( \varepsilon_p/\varepsilon = 10^2 \), as functions of the volume fraction of particles \( c \) (no results for the effective bulk modulus \( \tilde{\kappa} \) are included since the overall response is nearly incompressible in this case). The rationale behind this choice of material parameters is that they are typical of many of the dielectric elastomer composites studied experimentally, such as for instance those of...
6. Sample results in the limit of small deformations and moderate electric fields

Liu et al. (2013), where the elastomer is silicone rubber ($\mu = 0.22$ MPa, $\kappa = 1$ GPa, $\varepsilon = 3.2\varepsilon_0$) and the particles are made out of titania ($\mu_p = 110$ GPa, $\kappa_p = 220$ GPa, $\varepsilon_p = 114\varepsilon_0$). In these and subsequent figures, the solid lines are associated with the theoretical results (3.56), while the dashed lines and solid circles stand for corresponding results based on the analytical solution (4.23) for isotropic suspensions of polydisperse spherical particles — the so-called differential coated sphere assemblage (DCS) presented in Section 4.1 — and on FE simulations for isotropic suspensions of monodisperse spherical particles discussed in Section 5.1.

As discussed in Chapter 3, Figs. 6.2(a), (b), (c) illustrate that the theoretical results for the
iterative microstructure of Chapter 3 are in fairly good agreement with the results for suspensions of polydisperse and also monodisperse spherical particles, save quantitatively for the effective shear modulus $\tilde{\mu}$ when $c > 0.15$ and for the effective electrostrictive coefficient $\tilde{m}_K$ when $c > 0.05$. Another immediate observation from Figs. 6.2(a) and (b) is that the addition of stiff high-permittivity particles enhances both the stiffness and permittivity of the dielectric elastomer. Figure 6.2(c) shows that this is also the case for the electrostrictive coefficients $\tilde{m}_K$ and $\tilde{m}_J$. According to equation (6.1), and as expected on physical grounds, these trends set up a direct competition of effects for the overall electrostriction capabilities of the composite. To see which enhancement proves dominant, if the enhancement in stiffness (which makes the material less deformable) or that in permittivity and electrostrictive coefficients (which makes the material more prone to deform under the application of an electric field), we turn to examine the behavior of the ratio of electrostrictions $\overline{H}_{33}/H_{33}^a$ associated with the response of the composite under a uniaxial electric field; see Section 6.1.1.

Figure 6.2(d) shows plots of the ratio $\overline{H}_{33}/H_{33}^a$ as a function of the volume fraction of particles $c$ (no results for the ratio $\overline{H}_{11}/H_{11}^a$ are shown here since the overall near incompressibility of the composite implies that $\overline{H}_{11}/H_{11}^a \approx -1/2 \overline{H}_{33}/H_{33}^a$). The theoretical results indicate that $\overline{H}_{33}/H_{33}^a < 1$, that is, the addition of particles leads to a reduction in electrostriction. Physically, this entails that the enhancement in stiffness due to the addition of particles dominates over the enhancement in permittivity resulting in the filled elastomer undergoing less electrostriction than the unfilled elastomer when exposed to the same electric field. This behavior is in contrast to that displayed by the suspensions of spherical particles, which initially and up to about $c = 0.3$ exhibit an enhancement in electrostriction ($\overline{H}_{33}/H_{33}^a > 1$) before displaying a reduction ($\overline{H}_{33}/H_{33}^a < 1$).

While qualitatively different, all three sets of results in Fig. 6.2(d) agree in that the reduction or enhancement is quantitatively small, indeed $0.8 < \overline{H}_{33}/H_{33}^a < 1.15$ for the entire range of particle volume fractions considered, $0 \leq c \leq 0.4$. Such a difference in qualitative behavior but agreement in quantitative behavior among three different exact results for three different two-phase particulate isotropic microstructures suggest that the electrostriction capabilities of dielectric elastomers filled isotropically with stiff high-permittivity particles is highly sensitive to the details of the microstructure, but only in a qualitative manner. Quantitatively, moreover, they suggest that the enhancement in stiffness provided by the addition of filler particles essentially cancels out the enhancement in permittivity, so that there is ultimately little difference between the electrostriction capabilities of the unfilled and the filled elastomer (for particle volume fractions sufficiently away from percolation, of
Having gained insight into the theoretical results (3.56) for the case of stiff high-permittivity filler particles, we now deploy them to scrutinize two representative experiments: i) the experiments of Liu et al. (2013) for the electrostriction of a silicone (PDMS) elastomer filled with titania (TiO$_2$) particles and ii) the experiments of Huang et al. (2005) for the electrostriction of a polyurethane (PU) elastomer filled with semi-conductor copper phthalocyanine oligomer (o-CuPc) particles, coated by a polyacrylic acid (PAA), under the application of a uniaxial electric field.

The material parameters describing the elastic dielectric response of the PDMS elastomer utilized by Liu et al. (2013) are approximately given by $\mu = 0.22$ MPa, $\kappa = 1$ GPa, $\varepsilon = 3.2\varepsilon_0$, while those of the TiO$_2$ particles are $\mu_p = 110$ GPa, $\kappa_p = 220$ GPa, $\varepsilon_p = 114\varepsilon_0$. The particles were reported to be roughly spherical in shape, about 3 $\mu$m in average diameter, and spatially well dispersed throughout the PDMS matrix. Figure 6.3(a) shows the experimentally measured electrostriction $\mathbf{H}_{11}$ in the transverse direction for the pure PDMS elastomer (solid triangles) and the PDMS elastomer filled with a volume fraction $c = 0.082$ of TiO$_2$ particles (hollow triangles), as functions of the magnitude of the applied electric field $E$. The dotted and solid lines correspond to the theoretical predictions for the electrostriction of the unfilled ($c = 0$) and the filled ($c = 0.082$) PDMS elastomer, respectively, based on the result (6.4)$_1$ with (3.56). The theoretical prediction for the filled PDMS elastomer based on the analytical solution (4.23) for an isotropic suspension of polydisperse spherical particles is also included in the figure (dashed line) for comparison purposes.

On the other hand, the material parameters describing the elastic dielectric response of the PU elastomer utilized by Huang et al. (2005) are approximately given by $\mu = 9$ MPa, $\kappa = 5$ GPa, $\varepsilon = 8\varepsilon_0$, while those of the o-CuPc filler particles are $\mu_p = 1$ GPa, $\kappa_p = 100$ GPa, $\varepsilon_p = 10^4\varepsilon_0$. In this case too, the particles were roughly spherical in shape, about 40 nm in diameter, and spatially well dispersed throughout the PU matrix. The volume fraction of the particles was reported to be approximately $c = 0.073$. Figure 6.3(b) shows the experimentally measured electrostriction $\mathbf{H}_{33}$ in the thickness direction for the pure PU elastomer (solid triangles) and the filled PU elastomer (hollow triangles), as functions of the magnitude of the applied electric field $E$. Similar to Fig. 6.3(a), the dotted and solid lines correspond to the theoretical predictions (6.4)$_2$ with (3.56) for the pure ($c = 0$) and the filled ($c = 0.073$) PU elastomer, while the dashed line corresponds to the theoretical prediction for the filled PU elastomer based on the analytical solution (4.23).
6. Sample results in the limit of small deformations and moderate electric fields

It is apparent from Fig. 6.3(a) that the response of the filled PDMS elastomer exhibits a significant enhancement in electrostriction, in the order of 50% increase, when compared with the pure PDMS elastomer. In disaccord with this experimentally observed enhancement, as already discussed within the context of Fig. 6.2(d), the theoretical predictions show little change between the electrostriction of the pure and the filled PDMS elastomers. Fig. 6.3(b) shows an even more glaring difference between the experimental response of the filled PU elastomer, which exhibits about a 20-fold enhancement in electrostriction compared to the pure PU elastomer, and the theoretical predictions. This dramatic difference occurs consistently for the entire range of deformations $H_{33}$ and electric fields $E$ considered, including small values of $H_{33}$ and $E$ for which the asymptotic premise of “small” deformations and “moderate” electric fields — upon which the formulas (6.4) and (3.56) are based — is expected to be applicable. These disagreements will be dealt with in subsequent chapters.

6.1.3 Results for liquid-like particles with high permittivity

Next, we examine the case of dielectric elastomer composites wherein the filler particles are liquid-like, in the sense that they are characterized by nearly incompressible behavior and vanishingly small shear stiffness, and exhibit a higher permittivity than the elastomeric matrix. Such properties are
distinctive of many common fluids such as water and special types of alloys such as Galinstan. The practical interest in this type of dielectric elastomer composites is that increasing the content of their fillers increases the overall permittivity at the same time that it also increases the overall deformability (in contrast to the mechanically stiff particles considered in the preceding subsection) and thus has the potential to bestow the resulting composites with exceptionally enhanced electrostriction capabilities.

Figure 6.4(a), (b), (c) show results for the normalized effective shear modulus $\tilde{\mu}/\mu$, permittivity $\tilde{\varepsilon}/\varepsilon$, and electrostrictive coefficients $\tilde{m}_K/\varepsilon$, $-2\tilde{m}_J/\varepsilon$ of a dielectric elastomer composite comprised of a nearly incompressible elastomeric matrix with $\kappa/\mu = 10^3$ and liquid-like high-permittivity filler particles with $\mu_p/\mu = 10^{-2}$, $\kappa_p/\mu = 10^3$, and $\varepsilon_p/\varepsilon = 10^2$, as functions of the volume fraction of particles $c$. Figure 6.4(d) shows the associated ratio of electrostrictions $\Pi_{33}/\Pi_{33}^m$ also as a function of $c$. 
Figure 6.4: Overall electromechanical response of a nearly incompressible dielectric elastomer ($\kappa/\mu = 10^3$) filled with a random isotropic distribution of liquid-like high-permittivity spherical particles ($\mu_p/\mu = 10^{-2}$, $\kappa_p/\mu = 10^3$, $\varepsilon_p/\varepsilon = 10^2$). Results are shown for: (a) the normalized effective shear modulus $\tilde{\mu}/\mu$, (b) the normalized effective permittivity $\tilde{\varepsilon}/\varepsilon$, (c) the normalized effective electrostrictive coefficients $\tilde{m}_K/\varepsilon$, $-2\tilde{m}_J/\varepsilon$, and (d) the ratio of electrostrictions $\tilde{H}_{33}/H_{33}$, all as functions of the volume fraction of particles $c$. The solid lines in the plots correspond to the theoretical results (3.56). The dashed lines and solid circles stand for corresponding results based on the analytical solution (4.23) for an isotropic suspension of polydisperse spherical particles and on FE simulations for an isotropic suspension of monodisperse spherical particles.
Consistent with the preceding case for mechanically stiff particles, the theoretical results (3.56) are seen to be in fairly good agreement with the results for suspensions of polydisperse and monodisperse spherical particles, save quantitatively for the effective electrostrictive coefficient $\tilde{m}_K$. As expected on physical grounds, Figs. 6.4(a) and (b) confirm that the addition of liquid-like high-permittivity particles decreases the overall shear stiffness but increases the overall permittivity of the composite. Figure 6.4(c) shows that the effective electrostrictive coefficients $\tilde{m}_K$ and $\tilde{m}_J$ increase also with the addition of such fillers. This monotonic decrease in stiffness together with the increase in permittivity and electrostrictive coefficients entail that the electrostriction capabilities of the resulting dielectric elastomer composite are enhanced with the addition of fillers. This is precisely what is shown by Fig. 6.4(d), which illustrates not only that indeed $\tilde{H}_{33}/H_{33} > 1$ for all $c$ but also reveals that more than a 50% enhancement in electrostriction can be achieved with the addition of a moderate content of liquid-like high-permittivity particles. It would be interesting to explore these encouraging findings experimentally.

6.1.4 Results for porous dielectric elastomers

Finally, we consider the overall electromechanical response of porous dielectric elastomers made up of a dielectric elastomer containing a random isotropic distribution of vacuous spherical pores. Here, it is important to recognize that vacuous pores are mechanically softer at the same time that they exhibit lower permittivity than elastomers. Thus their addition results in an increase in overall deformability but also a decrease in overall permittivity setting up — similar to the case of stiff high-permittivity fillers — a direct competition of effects for the overall electrostriction capabilities of the composite.

Figure 6.5 presents the effect that the addition of vacuous pores ($\mu_p = 0, \kappa_p = 0, \varepsilon_p = \varepsilon_0$) has on the normalized effective shear modulus $\tilde{\mu}/\mu$, bulk modulus $\tilde{\kappa}/\kappa$, permittivity $\tilde{\varepsilon}/\varepsilon$, and electrostrictive coefficients $\tilde{m}_K/\varepsilon$, $-2\tilde{m}_J/\varepsilon$ to a nearly incompressible dielectric elastomer with bulk modulus $\kappa/\mu = 10^3$ and permittivity$^1$ $\varepsilon/\varepsilon_0 = 3$.

$^1$These values are typical of silicone rubber.
Figure 6.5: Overall electromechanical response of a nearly incompressible dielectric elastomer ($\kappa/\mu = 10^3$) with permittivity $\varepsilon = 3\varepsilon_0$ containing a random isotropic distribution of vacuous spherical pores ($\mu_p = 0$, $\kappa_p = 0$, $\varepsilon_p = \varepsilon_0$). Results are shown for: (a) the normalized effective shear and bulk moduli $\tilde{\mu}/\mu$ and $\tilde{\kappa}/\kappa$, (b) the normalized effective permittivity $\tilde{\varepsilon}/\varepsilon$, and (c)–(d) the normalized components of the effective electrostrictive tensor $\tilde{m}_K/\varepsilon$ and $-2\tilde{m}_J/\varepsilon$, all as functions of the volume fraction of pores $c$. The solid lines in the plots correspond to the theoretical results (3.56). The dashed lines and solid circles correspond to the analytical solution (4.23) for an isotropic suspension of polydisperse spherical particles and to FE simulations for an isotropic suspension of monodisperse spherical particles.

Similar to the two preceding types of dielectric elastomer composites with high-permittivity fillers, the theoretical results (3.56) exhibit good agreement with the results for suspensions of polydisperse and monodisperse spherical pores. Figures 6.5(a)–(b) illustrate that the isotropic addition of vacuous spherical pores reduces significantly the shear stiffness and permittivity of the dielectric elastomer and reduces drastically its volumetric stiffness leading to a highly compressible overall behavior. Figure 6.5(c) indicates that the electrostrictive coefficient $\tilde{m}_K$ is also reduced by increasing porosity. On the other hand, Fig. 6.5(d) shows that the electrostrictive coefficient $\tilde{m}_J$ is initially decreased but subsequently increased with the further addition of pores. As anticipated above, such variations of the effective material constants yield a direct competition of effects for the overall electrostriction
Figure 6.6: Electrostriction of a nearly incompressible dielectric elastomer ($\kappa/\mu = 10^3$) with permittivity $\varepsilon = 3\varepsilon_0$ containing a random isotropic distribution of vacuous spherical pores ($\mu_p = 0$, $\kappa_p = 0$, $\varepsilon_p = \varepsilon_0$). Parts (a) and (b) show, respectively, the ratios of electrostrictions in the transverse and thickness directions, $H_{11}/H_{11}^m$ and $H_{33}/H_{33}^m$, as functions of the volume fraction of pores $c$. The solid lines in the plots correspond to expressions (6.6) based on the theoretical results (3.56). The dashed lines and solid circles stand for corresponding results based on the analytical solution (4.23) for an isotropic suspension of polydisperse spherical particles and on FE simulations for an isotropic suspension of monodisperse spherical particles.

Figure 6.6 shows the electrostriction ratios (6.6) for a porous dielectric elastomer with the same matrix and pore material properties as those considered in Fig. 6.5. Interestingly, the ratio of electrostrictions $H_{11}/H_{11}^p$ transverse to the applied electric field is noticeably enhanced ($H_{11}/H_{11}^p > 1$) with the addition of pores. At the same time, the ratio of electrostrictions $H_{33}/H_{33}^p$ in the direction of the applied electric field is also significantly and monotonically enhanced ($H_{33}/H_{33}^p > 1$). These results reveal that when isotropically adding vacuous spherical pores to (incompressible or) nearly incompressible dielectric elastomers, the reduction in stiffness dominates over the reduction in permittivity resulting in an enhancement of the electrostriction capabilities of these composite materials. Akin to the preceding results for dielectric elastomers filled with liquid-like high-permittivity particles, it would be interesting to explore these encouraging findings experimentally.

In summary, the above sample results have illustrated the capabilities of the general solutions (3.29), (3.30), (3.31) to provide quantitative insight into the overall electromechanical response of dielectric elastomer composites. They have also served to reveal that for the case of dielectric elastomers filled with a random isotropic distribution of stiff, high-permittivity or (semi-)conducting, roughly spherical particles — the case that has hitherto received most attention by the experimental community — interphasial phenomena may be crucial in understanding and exploiting the enhanced capabilities of the composite.
electrostriction that this promising class of materials are able to achieve. Furthermore, they have revealed that the study of dielectric elastomers filled with liquid-like high-permittivity or (semi-)conducting particles as well as porous dielectric elastomers may be worth pursuing.

6.2 Transversely isotropic dielectric elastomers filled with aligned fibers

We recall that a number of studies (see, e.g., Wang and Mark, 1990; Meddeb and Ounaies, 2012; Lu et al., 2012) have indicated that anisotropic fillers in the form of short (needle-like) and long (cylindrical) fibers can potentially lead to even larger enhancements than those endowed by spherical fillers. In addition, such a type of composites can be readily fabricated by means of modern synthesis/manufacturing processes (see, e.g., Lu et al. (2012); Park et al. (2012); López Jiménez and Pellegrino (2012)). Here, we restrict ourselves to examining dielectric elastomer composites wherein the fillers are aligned cylindrical fibers with circular cross section; in this case, \( \omega = \infty \) and the general formulae (3.49) reduce to expressions (3.66). Finally, we seek to identify what type of fillers may potentially lead to the enhancement of the overall elastic dielectric properties of dielectric elastomers.

6.2.1 Electrostriction

Similar to Section 6.1, we are concerned here with the common approach to characterize the electromechanical properties of dielectric elastomers (filled or unfilled), that is measuring their deformation while they are subjected to a uniaxial electric field in the absence of stress (see, e.g., Section 2.25 in Stratton (1941) and Pelrine et al. (1998)). Such an electrically induced deformation is usually referred to as *electrostriction*. From the constitute relation (2.26), it follows that, in the absence of stress when \( \vec{S} = 0 \), the macroscopic electrostriction \( \vec{H} \) induced in a dielectric elastomer composite by a uniform macroscopic electric field \( \vec{E} \) is formally given by

\[
\vec{H} = -\vec{\mathcal{L}}^{-1}\vec{\mathcal{M}} \vec{E} \otimes \vec{E}.
\] (6.7)
Now, exploiting the fact that the effective modulus of elasticity here is of the transversely isotropic form (3.39), its inverse \( \tilde{L}^{-1} \) can be conveniently written as (Walpole, 1981)

\[
\tilde{L}^{-1} = \frac{\tilde{d}_L}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \mathcal{E}^{(1)} + \frac{\tilde{c}_L}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \mathcal{E}^{(2)} + \frac{1}{\tilde{e}_L} \mathcal{E}^{(3)} + \frac{1}{\tilde{f}_L} \mathcal{E}^{(4)} - \frac{\tilde{g}_L}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \left( \mathcal{E}^{(5)} + \mathcal{E}^{(6)} \right),
\]

where it is recalled that the tensors \( \mathcal{E}^{(1)} \) through \( \mathcal{E}^{(6)} \) are defined by relations (3.40). Upon direct use of this expression, the expression (3.39) for the effective electrostrictive tensor \( \tilde{\mathbf{M}} \), and some algebraic manipulation, the electrostriction (6.7) can be written more explicitly as

\[
\tilde{H} = - \left[ \left( \frac{\tilde{d}_L \tilde{c}_M - 2 \tilde{g}_L \tilde{h}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \mathcal{E}^{(1)} + \left( \frac{\tilde{c}_L \tilde{d}_M - 2 \tilde{g}_L \tilde{h}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \mathcal{E}^{(2)} + \frac{\tilde{e}_M}{\tilde{e}_L} \mathcal{E}^{(3)} + \frac{\tilde{f}_M}{\tilde{f}_L} \mathcal{E}^{(4)} \left( \frac{\tilde{c}_M \tilde{d}_M - \tilde{g}_M \tilde{d}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \mathcal{E}^{(5)} + \left( \frac{\tilde{d}_L \tilde{h}_M - \tilde{g}_L \tilde{d}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \mathcal{E}^{(6)} \right] \tilde{E} \otimes \tilde{E}.
\]

It is of note that this relation depends on all five effective elastic constants (\( \tilde{c}_L, \tilde{d}_L, \tilde{e}_L, \tilde{f}_L, \tilde{g}_L \)) and all six effective electrostrictive constants (\( \tilde{c}_M, \tilde{d}_M, \tilde{e}_M, \tilde{f}_M, \tilde{g}_M, \tilde{h}_M \)). It is also worth remarking that when the applied electric field \( \tilde{E} \) is aligned with the axis of symmetry \( \mathbf{N} (\tilde{E} = \tilde{E} \mathbf{N}) \) or orthogonal to it (\( \mathbf{N} \cdot \tilde{E} = 0 \)), the electrostriction (6.9) reduces to

\[
\tilde{H} = - \left( \frac{\tilde{d}_L \tilde{h}_M - \tilde{g}_L \tilde{d}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \tilde{E}^2 (\mathbf{I} - \mathbf{N} \otimes \mathbf{N}) - \left( \frac{\tilde{c}_L \tilde{d}_M - 2 \tilde{g}_L \tilde{h}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \tilde{E}^2 \mathbf{N} \otimes \mathbf{N}
\]

for the former and to

\[
\tilde{H} = \frac{1}{2} \left( \frac{\tilde{e}_M}{\tilde{e}_L} - \frac{\tilde{d}_L \tilde{c}_M - 2 \tilde{g}_L \tilde{g}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \tilde{E}^2 (\mathbf{I} - \mathbf{N} \otimes \mathbf{N}) - \left( \frac{\tilde{c}_L \tilde{g}_M - \tilde{g}_L \tilde{c}_M}{\tilde{c}_L \tilde{d}_L - 2 \tilde{g}_L} \right) \tilde{E}^2 \mathbf{N} \otimes \mathbf{N} - \frac{\tilde{e}_M}{\tilde{e}_L} \tilde{E} \otimes \tilde{E}
\]

for the latter. As opposed to the general result (6.9), the “aligned” electrostriction (6.10) depends only on three effective elastic constants (\( \tilde{c}_L, \tilde{d}_L, \tilde{g}_L \)) and two effective electrostrictive constants (\( \tilde{d}_M, \tilde{h}_M \)). Similarly, the “orthogonal” electrostriction (6.11) depends only on four effective elastic constants (\( \tilde{c}_L, \tilde{d}_L, \tilde{e}_L, \tilde{g}_L \)) and three effective electrostrictive constants (\( \tilde{c}_M, \tilde{e}_M, \tilde{g}_M \)).

In addition to their theoretical value in providing a rigorous analytical solution for a fundamental nonlinear coupled problem, the thirteen effective electromechanical constants (3.49) provide a formidable tool to gain insight into how the addition of anisotropic particles may enhance the electromechanical properties of dielectric elastomers. In this section, for illustration purposes, we seek to gain some of that insight via sample results.
6. Sample results in the limit of small deformations and moderate electric fields

Figure 6.7: Schematic of (a) the undeformed and (b) the deformed configurations of a dielectric elastomer filled with a transversely isotropic distribution of cylindrical fibers ($N = e_3$) with circular cross section subjected to a Lagrangian electric field of magnitude $E = \Phi/L^2$ in the orthogonal direction to the fibers $e_2$. The electrostriction $H$ undergone by the composite is described by equations (6.12)–(6.13) with (3.66).

With the objective of presenting results that are directly relatable to standard experimental measurements, we further restrict our attention to examining the electrostriction of these composites when subjected to an electric field that is orthogonal to the fibers (this involves only a subset of the thirteen effective electromechanical constants that characterize their electromechanical behavior). As shown schematically by Fig. 6.7, we choose the $e_3$ axis of the laboratory frame of reference to coincide with the direction of the fibers $N$, while the $e_2$ axis is chosen to coincide with the direction of the applied electric field $E = \mathbf{E} \cdot e_2$. Given this choice of frame of reference, the electrostriction $H$ in the composite takes the simple diagonal form (cf. equation (6.11))

$$ H = \overline{H}_{11} e_1 \otimes e_1 + \overline{H}_{22} e_2 \otimes e_2 + \overline{H}_{33} e_3 \otimes e_3 $$

(6.12)

with

$$ \overline{H}_{11} = \left( \frac{\tilde{e}_M}{\tilde{e}_L} - \frac{\tilde{d}_L \tilde{c}_M - 2\tilde{g}_L \tilde{g}_M}{\tilde{c}_L \tilde{d}_L - 2\tilde{g}_L^2} \right) E^2 $$

$$ \overline{H}_{22} = - \left( \frac{\tilde{e}_M}{\tilde{e}_L} + \frac{\tilde{d}_L \tilde{c}_M - 2\tilde{g}_L \tilde{g}_M}{\tilde{c}_L \tilde{d}_L - 2\tilde{g}_L^2} \right) \frac{E^2}{2} $$

$$ \overline{H}_{33} = \frac{\tilde{g}_L \tilde{c}_M - \tilde{c}_L \tilde{g}_M}{\tilde{c}_L \tilde{d}_L - 2\tilde{g}_L^2} E^2 $$

(6.13)

where it is recalled that, for the case of cylindrical fibers of interest here, the effective electromechanical constants $\tilde{c}_L, \tilde{d}_L, \tilde{e}_L, \tilde{g}_L, \tilde{c}_M, \tilde{e}_M, \tilde{g}_M$ are given explicitly by expressions (3.66).

In the next three subsections, we present and discuss numerical results for the electrostriction components (6.13) for dielectric elastomer composites wherein the elastomer\(^2\) has permittivity $\varepsilon = 3.2\varepsilon_0$ and Lamé constant $\lambda = \infty$, while the fibers exhibit various permittivities and mechanical

\(^2\)These values are representative of silicone rubber.
behaviors ranging from stiff, to liquid-like, to vacuous. To better illustrate the effect that the addition of fibers has on the electrostriction performance of elastomers, the results are presented in terms of the ratios $\Phi_{11}/\Phi_{11}^m$, $\Phi_{22}/\Phi_{22}^m$, $\Phi_{33}/\Phi_{33}^m$, where

$$H_{11}^m = H_{33}^m = \frac{\varepsilon}{6\mu}E^2$$
$$H_{22}^m = -\frac{\varepsilon}{3\mu}E^2$$

stand for the components of the electrostriction that the underlying elastomer would undergo in the absence of fibers.

To aid the discussion, in all of the results that follow we include comparisons with a separate exact analytical solution, as well as with full-field solutions constructed by means of the finite-element (FE) method. The analytical solution corresponds to a dielectric elastomer filled with a special type of transversely isotropic distribution of cylindrical fibers — the so-called differential coated cylinder (DCC) assemblage presented in Section 4.2 — with circular cross sections that are polydisperse in size. On the other hand, the full-field simulations correspond to a microstructure with monodisperse fibers presented in Section 5.2.

### 6.2.2 Results for stiff fibers

We begin by examining dielectric elastomer composites with fibers that are mechanically stiff. For definiteness, given that most materials commonly utilized as fibers (e.g., carbon) are much stiffer than elastomers, we take the fibers to be rigid, $\mu_p = \lambda_p = \infty$. Due to the overall rigidity in the direction of the fibers and the overall incompressibility of the resulting composite, the electrostriction components (6.13), normalized by the matrix electrostriction components (6.14), take the particularly simple form

$$\Phi_{11}/\Phi_{11}^m = \frac{3(1-c)\left[8\varepsilon_p + (2-c^2-c)(\varepsilon_p-\varepsilon)^2\right]}{4(1+c)(1+c)\varepsilon + (1-c)\varepsilon_p^2}$$, 

$$\Phi_{22}/\Phi_{22}^m = \frac{1}{2}\Phi_{11}^m$$, 

$$\Phi_{33}/\Phi_{33}^m = 0.$$  (6.15)

Figure 6.8 shows plots of the electrostriction ratio $\Phi_{11}/\Phi_{11}^m$ for fibers with two different permittivities: $\varepsilon_p = 4\varepsilon_0$ and $10^4\varepsilon_0$. The former value corresponds to the permittivity of Nylon, while the latter can be viewed as the permittivity of carbon. Both of these materials have been widely utilized as reinforcing fibers in elastomers (see, e.g., (Lu et al., 2012; López Jiménez and Pellegrino, 2012)). The results are plotted as functions of the volume fraction of fibers $c$. In this and subsequent figures, the solid line, referred to as theory, stands for the ratio of electrostrictions based on the effective electromechanical constants (3.66). On the other hand, the dashed line stands for the response of dielectric elastomer composites with the polydisperse DCC microstructure discussed in Section 4.2,
while the solid circles correspond to the FE simulations of dielectric elastomer composites with the monodisperse microstructure discussed in Section 5.2.

A key observation from Fig. 6.8 is that an infinitesimal addition of rigid fibers generates a 50% enhancement in electrostriction in the transverse direction to the fibers, namely, $\overline{H}_{11}/H_{m11}$ for $c = 0^+$. This abrupt enhancement is independent of the permittivity of the fibers, $\varepsilon_p$, and solely due to their rigidity, which constrains the composite to deform only in the transverse $e_1$-$e_2$ plane (see Fig. 6.7). As the content of fibers increases, the electrostriction enhancement in the elastomer with the low-permittivity fibers ($\varepsilon_p = 4\varepsilon_0$) monotonically decreases, vanishing at a fiber volume fraction of about $c = 0.2$. The further increase in the content of fibers beyond that point leads to a reduction in electrostriction ($\overline{H}_{11}/H_{m11} < 1$). The electrostriction of the elastomer with the high-permittivity fibers ($\varepsilon_p = 10^4\varepsilon_0$) exhibits similar trends, but its reduction occurs at a much slower rate. In fact, even at the relatively large volume fraction of $c = 0.4$, the electrostriction enhancement remains about 30%.

To illustrate the particular mechanical and anisotropic nature of the above-described enhancement, it proves helpful to consider the ratios $\overline{H}_{11}/H_{m11}$ and $\overline{H}_{22}/H_{m22}$, where

$$\hat{H}_{11}^n = -\hat{H}_{22}^n = \frac{\varepsilon}{4\mu} E^n_{33} \quad \text{and} \quad \hat{H}_{33}^n = 0$$

(6.16)

stand for the components of the electrostriction that the unfilled elastomer would undergo under
$e_1$-$e_2$ plane-strain conditions. These ratios read explicitly as

$$\frac{\Pi_{11}}{H_{11}^n} = \frac{\Pi_{22}}{H_{22}^n} = 1 - \frac{c[(3 + c)\varepsilon + (1 - c)\varepsilon_p]^2}{2(1 + c)(1 + c)\varepsilon + (1 - c)\varepsilon_p]^2}. \quad (6.17)$$

Similar to Fig. 6.8, Fig. 6.9 shows plots of the ratio $\Pi_{11}/H_{11}^n$ for fibers with the two different permittivities $\varepsilon_p = 4\varepsilon_0$ and $10^4\varepsilon_0$ in terms of the volume fraction of fibers $c$. Contrary to Fig. 6.8, as expected on physical grounds, an infinitesimal addition of rigid fibers does not yield any enhancement of the plane-strain electrostriction as $\Pi_{11}/H_{11}^n = 1$ for $c = 0^+$. Further addition of fibers leads to a significant reduction in the plane-strain electrostriction for the case of low-permittivity ($\varepsilon_p = 4\varepsilon_0$) fibers, and to a tenuous reduction for the case of high-permittivity ($\varepsilon_p = 10^4\varepsilon_0$) fibers, at least for the range of volume fractions considered. This behavior of a 2D isotropic distribution of rigid circular disks is analogous to the response of its 3D counterpart, an isotropic distribution of rigid spherical particles, presented in Section 6.1.2.

Figure 6.9: Electrostriction ratio $\Pi_{11}/H_{11}^n$ under $e_1$-$e_2$ plane-strain conditions of an incompressible ($\lambda = \infty$) dielectric elastomer with permittivity $\varepsilon = 3.2\varepsilon_0$ filled with a transversely isotropic distribution of rigid ($\mu_p = \lambda_p = \infty$) cylindrical fibers with circular cross section; see Fig. 6.7. Results are shown for fibers with permittivities $\varepsilon_p = 4\varepsilon_0$ and $10^4\varepsilon_0$, as functions of the volume fraction of fibers $c$. The solid line corresponds to the theoretical result (6.17). The dashed line corresponds to the response of a DCC assemblage of polydisperse fibers (see Section 4.2), while the solid circles correspond to FE simulations of a dielectric elastomer composite with monodisperse fibers (see Section 5.2).

Another key observation from Figs. 6.8–6.9 is the close agreement between the theoretical predictions (6.15), (6.17) and the DCC and FE results. This agreement among three different exact results for three different microstructures suggests that the electrostriction capabilities of dielectric elastomers filled with transversely isotropic distributions of cylindrical fibers is fairly insensitive to “higher-order” microstructural details such as the cross sectional shape and the size of the fibers, so long as the microstructure is sufficiently far away from its percolation limit.
6.2.3 Results for liquid-like fibers

Next, we consider dielectric elastomer composites with fibers that are liquid-like, in the sense that they are incompressible, \( \lambda_p = \infty \), and exhibit vanishingly small shear resistance, \( \mu_p = 0 \). Fibers with such properties could be made, for instance, by filling manufactured cylindrical cavities in the dielectric elastomer of interest with common fluids such as water or with eutectic alloys such as Galinstan (Park et al., 2012; Wang et al., 2012; Fassler and Majidi, 2015). As already alluded to in Section 6.1.3, an attractive feature of this type of dielectric elastomer composites is that increasing their content of fibers can increase their overall permittivity (if \( \varepsilon_p > \varepsilon \)) at the same time that it also increases their overall deformability (since \( \mu_p < \mu \)) and thus has the potential to bestow the resulting composites with exceptionally enhanced electrostriction capabilities. The electrostriction components (6.13), when normalized by the matrix electrostriction components (6.14), reduce in this case to

\[
\frac{H_{11}}{H_{m11}} = 1 + \frac{c(9 - c)}{4(1 - c)^2} + \frac{c\varepsilon[\varepsilon + (3c^2 + 5)(\varepsilon_p - \varepsilon) - c(\varepsilon + 8\varepsilon_p)]}{(1 - c)^2[(1 + c)\varepsilon + (1 - c)\varepsilon_p]^2},
\]

\[
\frac{H_{22}}{H_{m22}} = \frac{1}{2} \left( \frac{H_{11}}{H_{m11}} + \frac{H_{33}}{H_{m33}} \right),
\]

\[
\frac{H_{33}}{H_{m33}} = 1 + \frac{c[(3 - c)\varepsilon_p - (1 - c)\varepsilon]}{(1 - c)[(1 + c)\varepsilon + (1 - c)\varepsilon_p]].
\]

(6.18)

Figure 6.10 shows results for the electrostriction ratios \( H_{11}/H_{m11} \) and \( H_{33}/H_{m33} \) for fibers with permittivity \( \varepsilon_p = 10^2 \varepsilon_0 \), a value representative of the permittivity of water, as functions of the volume fraction of fibers \( c \). As expected, the enhancement in both electrostriction ratios with the addition of such liquid-like fibers is very significant, exceeding 200\% for fiber volume fractions \( c > 0.3 \). Consistent with the previous case of stiff fibers, the theoretical results (6.18) are seen to be in fairly good agreement with the DCC and FE results.
6.2.4 Results for vacuous cylindrical pores

As a final set of sample results, we consider the electrostriction of dielectric elastomer composites containing aligned cylindrical vacuous cavities or pores, \( \lambda_p = \mu_p = 0, \varepsilon_p = \varepsilon_0 \). For this choice of “fillers”, expressions (6.13), again, when normalized by (6.14), specialize to

\[
\begin{align*}
\frac{H_{11}}{H_{11}^m} &= 1 + \frac{(21 - c)c}{4(1 - c)^2} - \frac{c\varepsilon[\varepsilon_0 + (10 + 9c)\varepsilon + c(8 - 9c)(\varepsilon_0 - \varepsilon)]}{(1 - c)^2[(1 + c)\varepsilon + (1 - c)\varepsilon_0]^2}, \\
\frac{H_{22}}{H_{22}^m} &= 1 + \frac{(9 - 5c)c}{8(1 - c)^2} + \frac{c\varepsilon[(1 - c)(3c + 7)\varepsilon_0 - (2 - c)(1 + 3c)\varepsilon]}{2(1 - c)^2[(1 + c)\varepsilon + (1 - c)\varepsilon_0]^2}, \\
\frac{H_{33}}{H_{33}^m} &= 1 - \frac{c[(1 - c)\varepsilon - (3 - c)\varepsilon_0]}{(1 - c)[(1 + c)\varepsilon + (1 - c)\varepsilon_0]}.
\end{align*}
\]

(6.19)

Figure 6.11 shows plots of these three electrostriction ratios as functions of the volume fraction of pores \( c \). While the electrostriction ratio \( \frac{H_{22}}{H_{22}^m} \) associated with the direction of the applied electric field (see Fig. 6.7) increases rather significantly with the addition of pores, the electrostriction ratio \( \frac{H_{33}}{H_{33}^m} \) slowly departs from unity, and \( \frac{H_{11}}{H_{11}^m} \) slightly reduces before increasing and reaching an enhancement for fiber volume fraction \( c > 0.25 \). These results reveal that the addition of cylindrical pores to dielectric elastomers introduces a strong anisotropy in their electrostriction response, with enhanced performance in the transverse direction to the pores. Similar to the two previous cases of stiff and liquid-like fibers, Fig. 6.11 shows that the theoretical results (6.19) for cylindrical pores
agree quite closely with the DCC and FE results sufficiently away from percolation.

Figure 6.11: Electrostriction ratios $\frac{H_{11}}{H_{m11}}$, $\frac{H_{22}}{H_{m22}}$, $\frac{H_{33}}{H_{m33}}$ of an incompressible ($\lambda = \infty$) dielectric elastomer with permittivity $\varepsilon = 3\varepsilon_0$ containing a transversely isotropic distribution of vacuous ($\mu_p = \lambda_p = \infty$, $\varepsilon_p = \varepsilon_0$) cylindrical pores with circular cross section, as functions of the volume fraction of pores $c$; see Fig. 6.7. The solid line corresponds to the theoretical results (6.19). The dashed line corresponds to the response of a DCC assemblage of polydisperse cylindrical pores, while the solid circles correspond to FE simulations of a dielectric elastomer composite with monodisperse cylindrical pores.

In summary, the above sample theoretical results indicate that the addition of anisotropic fillers — in this case, aligned cylindrical fibers — to dielectric elastomers can lead to substantial enhancements in the electrostriction capabilities of this emerging class of soft active materials. More specifically, the results indicate that *liquid-like* and even *porous anisotropic fillers* have the potential to lead to significantly more pronounced enhancements than those endowed by *mechanically stiff anisotropic fillers*. It would be interesting to explore these findings experimentally.
Nonlinear electroelastic deformations of dielectric elastomer composites: ideal elastic dielectrics

Naviguer: c’est accepter les contraintes que l’on a choisies. C’est un privilège. La plupart des humains subissent les obligations que la vie leur a imposées.

– Éric Tabarly, Mémoires du large, 1998

The four preceding chapters have addressed the macroscopic elastic dielectric response of dielectric elastomer composites in the limit of small deformations and moderate electric fields. In this and subsequent chapter, we address their response under finite deformations and finite electric fields.

We begin here by considering the fundamental case when the matrix and the particles are both ideal elastic dielectrics. Further, we restrict our attention to the practically prominent case of dielectric elastomer composites with isotropic microstructures.

**Isotropic ideal elastic dielectric composites.** Specifically, we seek to generate solutions for the effective free energy (2.18) for the case of dielectric elastomer composites wherein the distribution of filler particles is isotropic and both the elastomeric matrix and the filler particles are ideal elastic dielectrics. Namely, the elastic dielectric behaviors of the matrix and particles are characterized by
the following free-energy functions

\[
W^{(1)}(F, E) = \begin{cases} 
\frac{\mu}{2} [I_1 - 3] - \frac{\varepsilon}{2} I_5^E & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]

and

\[
W^{(2)}(F, E) = \begin{cases} 
\frac{\mu_p}{2} [I_1 - 3] - \frac{\varepsilon_p}{2} I_5^E & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]  

(7.1)

where the standard notation \( I_1 = F \cdot F \), \( I_5^E = F^{-T}E \cdot F^{-T}E \) has been employed, \( \mu, \mu_p \) denote the initial shear moduli of the matrix and the particles, and \( \varepsilon, \varepsilon_p \) stand for their respective initial permittivities; basic physical considerations dictate that \( \mu, \mu_p > 0 \) and \( \varepsilon, \varepsilon_p \geq \varepsilon_0 \), where, again, \( \varepsilon_0 \) denotes the permittivity of vacuum. Here, it is appropriate to remark that the choice (7.1)_1 is the simplest valid prototype for dielectric elastomers. This is because their elasticity can be approximated as Gaussian up to moderate levels of deformation and their polarization \( p = FD - \varepsilon_0 F^{-T}E \) is rather insensitive to the state of deformation and varies roughly linearly with the applied Eulerian electric field \( e = F^{-T}E \) (see, e.g., Kofod et al., 2003; Wissler and Mazza, 2007; Di Lillo et al., 2011). We also note that the choice (7.1)_2 includes extremal behaviors of notable relevance in applications such as rigid conducting particles, corresponding to the choice \( \mu_p = +\infty \) and \( \varepsilon_p = +\infty \), and liquid conducting particles, corresponding to \( \mu_p = 0 \) and \( \varepsilon_p = +\infty \) (see, e.g., Huang et al., 2005; Fassler and Majidi, 2015).

In light of the assumed isotropy of the microstructure and the constitutive isotropy and incompressibility of the matrix material and filler particles (7.1), the macroscopic elastic dielectric response of the resulting dielectric elastomer composite is itself isotropic and incompressible. As a result, its effective free-energy function \( \overline{W} \) given by (2.18) only depends on the macroscopic deformation gradient \( F \) and macroscopic Lagrangian electric field \( E \) through five invariants and becomes unbounded for non-isochoric deformations when \( J = \det F \neq 1 \). With a slight abuse of notation, we write

\[
\overline{W}(F, E, c) = \begin{cases} 
\overline{W}(\overline{I}_1, \overline{I}_2, \overline{I}_4^E, \overline{I}_5^E, \overline{I}_6^E, c) & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]  

(7.2)

in terms of the five standard invariants

\[
\begin{align*}
\overline{I}_1 &= F \cdot F, & \overline{I}_2 &= F^{-T} \cdot F^{-T}, & \overline{I}_4^E &= E \cdot E, \\
\overline{I}_5^E &= F^{-T}E \cdot F^{-T}E, & \overline{I}_6^E &= F^{-1}F^{-T}E \cdot F^{-1}F^{-T}E.
\end{align*}
\]  

(7.3)

We shall also find it useful to express \( \overline{W} \) alternatively in terms of two of the singular values of \( F \), \( \overline{\lambda}_1 \) and \( \overline{\lambda}_2 \) say, with the third one \( \overline{\lambda}_3 = (\overline{\lambda}_1 \overline{\lambda}_2)^{-1} \), and the three components of the macroscopic
Lagrangian electric field, \( \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3 \), with respect to an arbitrary frame of reference. With a slight abuse of notation as in (7.2), we write

\[
W(\mathbf{F}, \mathbf{E}, c) = \begin{cases} 
W(\mathbf{X}_1, \mathbf{X}_2, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3, c) & \text{if } \mathbf{X}_3 = (\mathbf{X}_1 \mathbf{X}_2)^{-1} \\
+\infty & \text{otherwise}
\end{cases}
\]  

(7.4)

and note the symmetries

\[
W(\mathbf{X}_1, \mathbf{X}_2, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3, c) = W(\mathbf{X}_2, \mathbf{X}_1, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3, c) = W(\mathbf{X}_1, (\mathbf{X}_1 \mathbf{X}_2)^{-1}, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3, c) = W((\mathbf{X}_1 \mathbf{X}_2)^{-1}, \mathbf{X}_1, \mathbf{E}_3, \mathbf{E}_1, \mathbf{E}_2, c) = W((\mathbf{X}_1 \mathbf{X}_2)^{-1}, \mathbf{X}_2, \mathbf{E}_3, \mathbf{E}_1, \mathbf{E}_2, c) = W(\mathbf{X}_2, (\mathbf{X}_1 \mathbf{X}_2)^{-1}, \mathbf{E}_2, \mathbf{E}_3, \mathbf{E}_1, c).
\]

**Power series solution about the ground state** \( \mathbf{F} = \mathbf{I} \) and \( \mathbf{E} = 0 \). For later reference, we note here that the effective free-energy function \( W \) of (the majority of) isotropic ideal elastic dielectric composites is expected to admit a power series solution about the ground state \( \mathbf{F} = \mathbf{I} \) and \( \mathbf{E} = 0 \). Expressing such a solution in terms of the variables \( \mathbf{X}_1, \mathbf{X}_2, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3 \), the finite branch of (7.4) reads as

\[
W(\mathbf{X}_1, \mathbf{X}_2, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3, c) = \sum_{m,n,p,q,r=0}^{\infty} k_{mnopq} (\lambda_1 - 1)^m (\lambda_2 - 1)^n \mathbf{E}_1^p \mathbf{E}_2^q \mathbf{E}_3^r,
\]

(7.5)

where the coefficients \( k_{mnopq} \) are functions of the microstructure, as characterized by the indicator function \( \theta \) in (2.13), as well as of the material parameters \( \mu, \mu_p, \varepsilon, \varepsilon_p \).

Given the properties \( W^{(1)}(\mathbf{F}, -\mathbf{E}) = W^{(1)}(\mathbf{F}, \mathbf{E}) \) and \( W^{(2)}(\mathbf{F}, -\mathbf{E}) = W^{(2)}(\mathbf{F}, \mathbf{E}) \) of the local free energies (7.1), it follows from the definition (2.18) of \( W \) that the electromechanical coupling of the overall response of the composite is even, namely,

\[
W(\mathbf{X}_1, \mathbf{X}_2, -\mathbf{E}_1, -\mathbf{E}_2, -\mathbf{E}_3, c) = W(\mathbf{X}_1, \mathbf{X}_2, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3, c).
\]

This implies that the coefficients \( k_{mnopq} = 0 \) in (7.5) when \( p+q+r = 2n+1 \) with \( n \in \mathbb{N} \). Moreover, given the additional properties \( W^{(1)}(\mathbf{I}, 0) = W^{(2)}(\mathbf{I}, 0) = 0 \), it follows from the definition (2.18) that

\[
k_{00000} = k_{10000} = k_{01000} = k_{00100} = k_{00010} = k_{00001} = k_{10020} = k_{01200} = 0,
\]

\[
k_{20000} = k_{02000} = k_{11000} = 2\mu,
\]

\[
k_{00200} = k_{00020} = k_{00002} = -\frac{\varepsilon}{2},
\]

\[
k_{10200} = -k_{10020} = k_{01020} = -k_{01002} = \tilde{m}_K,
\]

(7.6)

and hence that, in the neighborhood of the ground state,

\[
W(\mathbf{X}_1, \mathbf{X}_2, \mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3, c) = 2\mu \left[ (\mathbf{X}_1 - 1)^2 + (\mathbf{X}_2 - 1)^2 + (\mathbf{X}_1 - 1)(\mathbf{X}_2 - 1) \right] - \frac{\varepsilon}{2} \left[ \mathbf{E}_1^2 + \mathbf{E}_2^2 + \mathbf{E}_3^2 \right] + \tilde{m}_K \left[ (\mathbf{X}_1 - 1)(\mathbf{E}_1^2 - \mathbf{E}_3^2) + (\mathbf{X}_2 - 1)(\mathbf{E}_2^2 - \mathbf{E}_3^2) \right].
\]

(7.7)
plus higher-order correction terms. In the above expressions, \( \bar{\mu} \), \( \bar{\varepsilon} \), \( \bar{m}_K \) denote the effective shear modulus, effective permittivity, and effective electrostrictive constant that characterize the electromechanical response of the composite in the classical limit of small deformations and moderate electric fields; see Section 2.2.1. In the present context, these effective constants are given by the formulae

\[
\bar{\mu} = \frac{1}{5} \int_{\Omega} \mu(X) K_{ijklm} \Gamma_{mkl,n} dX,
\]

\[
\bar{\varepsilon} = \int_{\Omega} \varepsilon(X) \gamma_{m,m} dX,
\]

\[
\bar{m}_K = \frac{1}{5} \int_{\Omega} \varepsilon(X) K_{ijkl} \Gamma_{ijkl} \Gamma_{rsuv} \gamma_{u,k} \gamma_{v,l} dX,
\]

(7.8)

where \( \mu(X) = [1 - \theta(X)] \mu + \theta(X) \mu_p \), \( \varepsilon(X) = [1 - \theta(X)] \varepsilon + \theta(X) \varepsilon_p \), \( K_{ijkl} = \frac{1}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) - \frac{1}{3} \delta_{ij} \delta_{kl} \) with, again, \( \delta_{ij} \) denoting the Kronecker delta, the notation \( \cdot, \cdot \) represents partial differentiation with respect to the material point coordinate \( X_i \), and the tensor fields \( \Gamma \) and \( \gamma \) are implicitly defined as the solutions of the following uncoupled linear boundary value problems:

\[
\left\{ \begin{array}{l}
\left[ \mu(X) K_{ijklm} \Gamma_{mkl,n} + \frac{1}{2} \delta_{ij} \delta_{kl} \right]_{,l} = 0 \\
\Gamma_{mkl,m} = 0
\end{array} \right. 
\quad \text{for } X \in \Omega, \quad \Gamma_{ijkl} = \delta_{ik} X_l \quad \text{for } X \in \partial \Omega \quad (7.9)
\]

and

\[
\left[ \varepsilon(X) \gamma_{i,j} \right]_{,i} = 0 
\quad \text{for } X \in \Omega, \quad \gamma_{i} = X_{i} \quad \text{for } X \in \partial \Omega. \quad (7.10)
\]

### 7.1 A solution for an isotropic iterative microstructure

As already mentioned in Chapter 3, by means of a combination of iterative techniques, Lopez-Pamies (2014) constructed an exact solution for the variational problem (2.18) for a fairly general class of two-phase particulate microstructures wherein the particles are infinitely polydisperse in size; see Section 3.1. When specialized to isotropic distributions of filler particles and to matrix and filler particle behaviors characterized by the ideal elastic dielectric free energies (7.1), his result for the finite branch of the effective free-energy function \( \bar{W} \), expressed in terms of the variables...
\( \mathcal{W}(\lambda_1, \lambda_2, E_1, E_2, E_3, c) \) can be written as

\[
\frac{1}{2} \left( \lambda_1^2 + \lambda_2^2 + \frac{1}{\lambda_1 \lambda_2} - 3 \right) - \frac{\varepsilon}{2} \left[ \frac{E_1^2}{\lambda_1^2} + \frac{E_2^2}{\lambda_2^2} + \lambda_1 \lambda_2 E_3^2 \right].
\]

Here, the function \( U = U(\lambda_1, \lambda_2, E_1, E_2, E_3, c) \) is defined as the viscosity solution of the first order nonlinear pde

\[
\frac{\varepsilon}{2} \frac{\partial U}{\partial c} - U - \sum_{m,n,p,q,r=0}^{2} \alpha_{mnpq} \left( \frac{\partial U}{\partial \lambda_1} \right)^m \left( \frac{\partial U}{\partial \lambda_2} \right)^n \left( \frac{\partial U}{\partial E_1} \right)^p \left( \frac{\partial U}{\partial E_2} \right)^q \left( \frac{\partial U}{\partial E_3} \right)^r = 0
\]

subject to the initial condition

\[
U(\lambda_1, \lambda_2, E_1, E_2, E_3, 1) = \frac{1}{4} \left( \frac{\mu_p^2}{\mu} - 1 \right) \left[ \lambda_1^2 + \lambda_2^2 + \frac{1}{\lambda_1 \lambda_2} - 3 \right] + \frac{\varepsilon - \varepsilon_p}{4 \mu} \left[ \frac{E_1^2}{\lambda_1^2} + \frac{E_2^2}{\lambda_2^2} + \lambda_1 \lambda_2 E_3^2 \right],
\]

where the fifteen coefficients \( \alpha_{mnpq} \) in (7.12) are given by expressions (D.1) in Appendix D.1 due to their bulkiness. The pde (7.12) is a Hamilton-Jacobi (HJ) equation where, in the standard parlance in the study of this class of pdes (see, e.g., Benton, 1977), the volume fraction of particles \( c \) corresponds to the “time” variable and the five electromechanical loading variables \( \lambda_1, \lambda_2, E_1, E_2, E_3 \) correspond to the “space” variables. In spite of its nonlinear nature, its viscosity solution can be worked out in closed form in the asymptotic contexts of: i) small deformations and moderate electric fields and ii) infinitely large deformations. These asymptotic solutions are the subjects of the next two subsections. More generally, for arbitrary finite deformations and finite electric fields, the initial-value problem (7.12)–(7.13) does not appear to admit a closed-form solution. We present in Section 7.1.3 numerical solutions for it that are generated by means of a WENO finite-difference scheme.

7.1.1 The limit of small deformations and moderate electric fields

The asymptotic solution for the effective free-energy function of dielectric elastomer composites with the general iterative microstructure of Lopez-Pamies (2014) was derived in Chapter 3 in the limit of small deformations and moderate electric fields, in the present context, when \( \bar{\lambda}_1, \bar{\lambda}_2 \to 1 \) and \( E_1, E_2, E_3 \to 0 \). This solution includes as a special case the solution of interest here for isotropic...
ideal elastic dielectric composites defined by (7.11) with (7.12)–(7.13). When expressed in terms of the present notation, it reads as

\[
W(\lambda_1, \lambda_2, E_1, E_2, E_3, c) = \frac{6(1-c)\mu + 2(2 + 3c)\mu_p}{(3 + 2c)\mu + 2(1-c)\mu_p} \left[ (\lambda_1 - 1)^2 + (\lambda_2 - 1)^2 + (\lambda_1 - 1)(\lambda_2 - 1) \right] - \\
\frac{2(1-c)\varepsilon + (1+2c)\varepsilon_p \varepsilon}{2[(2+c)^2 + (1-c)\varepsilon_p]} \left[ E_1^2 + E_2^2 + E_3^2 \right] + \\
\left( \varepsilon + \frac{3c(1-c)(\varepsilon_p - \varepsilon)}{10[(2+c)\varepsilon + (1-c)\varepsilon_p]^2} \right) \left[ (\lambda_1 - 1)(E_1^2 - E_3^2) + (\lambda_2 - 1)(E_2^2 - E_3^2) \right]
\]

(7.14)

plus higher-order correction terms. This asymptotic result about the ground state is, of course, of the form (7.7), where the effective constants are given by

\[
\bar{\mu} = \frac{3(1-c)\mu + (2+3c)\mu_p}{(3+2c)\mu + 2(1-c)\mu_p}, \\
\bar{\varepsilon} = \frac{2(1-c)\varepsilon + (1+2c)\varepsilon_p \varepsilon}{(2+c)\varepsilon + (1-c)\varepsilon_p}, \\
\bar{m}_K = \varepsilon + \frac{3c(1-c)(\varepsilon_p - \varepsilon)}{10[(2+c)\varepsilon + (1-c)\varepsilon_p]^2} \left( 7\varepsilon_p + \frac{(23+7c)\varepsilon}{1-c} - \frac{15\varepsilon_p(\varepsilon_p - \varepsilon)}{(3+2c)\mu + 2(1-c)\mu_p} \right) \varepsilon.
\]

There are a number of different representations in terms of the set of five standard invariants (7.3) that are consistent with the asymptotic result (7.14). For reasons that will become apparent below, here, we spell out one such form:

\[
W(T_1, T_2, T_4^E, T_5^E, T_6^E, c) = \frac{\bar{\mu}}{2} [T_1 - 3] + \frac{\bar{m}_K - \bar{\varepsilon}}{2} T_4^E - \frac{\bar{m}_K}{2} T_5^E.
\]

(7.16)

Note that this representation is linear in the invariants \(T_1, T_4^E, T_5^E\) and independent of the two other invariants \(T_2, T_6^E\).

### 7.1.2 The limit of infinitely large deformations

In the limit of infinitely large deformations when \(\lambda_1 \to 0, +\infty\) and/or \(\lambda_2 \to 0, +\infty\), the solution for the effective free-energy function (7.11) with (7.12)–(7.13) can also be worked out in closed form. To avoid loss of continuity, the pertinent asymptotic analysis is summarized in Appendix D.2. The result can be compactly written in a single expression as

\[
W(\lambda_1, \lambda_2, E_1, E_2, E_3, c) = \frac{2(1-c)\mu + (1+2c)\mu_p}{2[(2+c)\mu + 2(1-c)\mu_p]} \left[ \frac{\lambda_1^2 + \lambda_2^2 + 1}{\lambda_1^2 \lambda_2^2} \right] - \\
\frac{\varepsilon_p}{2[c\varepsilon + (1-c)\varepsilon_p]} \left[ \frac{E_1^2}{\lambda_1^2} + \frac{E_2^2}{\lambda_2^2} + \lambda_1^2 \lambda_2^2 E_3 \right]
\]

(7.17)
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plus higher-order correction terms.

An obvious representation in terms of the set of invariants (7.3) that is consistent with the asymptotic result (7.17) is given by

\[ W(I_1, I_2, I_4^E, I_5^E, I_6^E, c) = \frac{[2(1 - c)\mu + (1 + 2c)\mu_p]}{2[(2 + c)\mu + 2(1 - c)\mu_p]}\mu I_1 - \frac{\varepsilon_p}{2[c\varepsilon + (1 - c)\varepsilon_p]}\varepsilon I_5^E. \]  

(7.18)

Much like the representation (7.16) for the limit of small deformations and moderate electric fields, the representation (7.18) for infinitely large deformations is also linear in the invariants \( I_1, I_4^E, I_5^E \), with the proportionality constant for \( I_4^E \) being zero, and independent of \( I_2, I_6^E \).

7.1.3 Finite deformations and finite electric fields

For arbitrary values of finite stretches \( \lambda_1, \lambda_2 \) and finite electric fields \( E_1, E_2, E_3 \), the HJ equation (7.12)–(7.13) for the function \( \overline{U} \) in the effective free energy (7.11) requires a numerical approach. In the sequel, we present a sample of such numerical solutions generated by means of a new scheme recently designed for this class of HJ pdes. As essential elements, we mention that the scheme employs a monotone numerical Hamiltonian (Crandall and Lions, 1984; Osher and Sethian, 1988) in combination with a fifth-order accurate WENO finite-difference discretization in the “space” variables \( \lambda_1, \lambda_2, E_1, E_2, E_3 \), including the grid points on the boundary of the domain of computation, and a fifth-order explicit Runge-Kutta “time” integration in \( c \); the interested reader is referred to Shu (2009) and references therein for a generic overview of WENO approaches. Basic technical details of the specific scheme that we employ here are provided in Appendix D.3.

To gain insight into the qualitative features of the effective free energy (7.11), we begin by plotting its value, normalized by the initial shear modulus of the matrix material \( \mu \), as a function of the stretch \( \lambda_1 \) and the normalized electric field component \( E_3/\sqrt{\mu/\varepsilon} \) for the experimentally prominent case of axisymmetric electromechanical loading with \( \lambda_2 = \lambda_1 \) and \( E_1 = E_2 = 0 \). Figure 7.1(a) shows the result for stiff high-permittivity particles with volume fraction \( c = 0.05 \) and material parameters \( \mu_p = 10^2 \mu \) and \( \varepsilon_p = 10^2 \varepsilon \), whereas Fig. 7.1(b) shows the corresponding result for liquid-like high-permittivity particles with volume fraction \( c = 0.15 \) and material parameters \( \mu_p = 10^{-1} \mu \) and \( \varepsilon_p = 10 \varepsilon \).
Further qualitative as well as quantitative insight into the effective free energy (7.11) can be gained by plotting its normalized value $W/\mu$ as a function of each of the five normalized invariants $\tilde{T}_1$, $\tilde{T}_2$, $\tilde{\varepsilon} \tilde{T}_4/\mu$, $\tilde{\varepsilon} \tilde{T}_5/\mu$, $\tilde{\varepsilon} \tilde{T}_6/\mu$ while keeping the remaining four invariants fixed. Figures 7.2 and 7.3 show such plots for the same volume fractions of particles and the same two sets of material parameters utilized in Fig. 7.1, namely, stiff high-permittivity particles with $\mu_p = 10^2\mu$ and $\varepsilon_p = 10^2\varepsilon$ at $c = 0.05$ and liquid-like high-permittivity particles with $\mu_p = 10^{-1}\mu$ and $\varepsilon_p = 10\varepsilon$ at $c = 0.15$. Note that fixing the values of four of the invariants $\tilde{T}_1$, $\tilde{T}_2$, $\tilde{\varepsilon} \tilde{T}_4/\mu$, $\tilde{\varepsilon} \tilde{T}_5/\mu$, $\tilde{\varepsilon} \tilde{T}_6/\mu$ restricts the range of physical values that the remaining invariant can take on. For example, for the fixed values $\tilde{T}_2 = 8.03$, $\tilde{\varepsilon} \tilde{T}_4/\mu = 0.61$, $\tilde{\varepsilon} \tilde{T}_5/\mu = 1.63$, $\tilde{\varepsilon} \tilde{T}_6/\mu = 10.41$ in Fig. 7.2(a), the range of physically allowable values of $\tilde{T}_1$ is $[5.75, 9.76]$. The results presented in Figs. 7.2 and 7.3 span the entire range of physically allowable values for each case that is presented.
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Figure 7.2: Plots of the effective free-energy function (7.11) of an ideal elastic dielectric, with initial shear modulus $\mu$ and initial permittivity $\varepsilon$, filled with stiff high-permittivity particles, with initial shear modulus $\mu_p = 10^2 \mu$ and initial permittivity $\varepsilon_p = 10^2 \varepsilon$, at volume fraction $c = 0.05$. Results are shown for the values of the normalized free energy $W/\mu$ in terms of each of the five normalized invariants $T_1, T_2, \varepsilon T_1^E/\mu, \varepsilon T_2^E/\mu, \varepsilon T_3^E/\mu$ for two sets of fixed values of the remaining four invariants. The solid lines (labeled “HJ Exact”) correspond to the numerical viscosity solution, while the dashed lines (labeled “HJ Approx.”) correspond to the closed-form approximation (7.26).
Figure 7.3: Plots of the effective free-energy function (7.11) of an ideal elastic dielectric, with initial shear modulus $\mu$ and initial permittivity $\varepsilon$, filled with liquid-like high-permittivity particles, with initial shear modulus $\mu_p = 10^{-1} \mu$ and initial permittivity $\varepsilon_p = 10 \varepsilon$, at volume fraction $c = 0.15$. Results are shown for the values of the normalized free energy $W/\mu$ in terms of each of the five normalized invariants $T_1$, $T_2$, $\varepsilon T_1^E/\mu$, $\varepsilon T_2^E/\mu$, $\varepsilon T_5^E/\mu$ for two sets of fixed values of the remaining four invariants. The solid lines (labeled “HJ Exact”) correspond to the numerical viscosity solution, while the dashed lines (labeled “HJ Approx.”) correspond to the closed-form approximation (7.26).
Much like in the two preceding asymptotic limits involving small and infinitely large deformations, the key observation from Figs. 7.2 and 7.3 is that, for finite deformations and finite electric fields too, the effective free-energy function (7.11) depends roughly linearly on the invariants $T_1, T_4^E, T_5^E$ and is roughly independent of $T_2, T_6^E$. The dependence on $T_4^E$ is much weaker than on $T_1$ and $T_5^E$. This is more so for the case of stiff high-permittivity particles than for liquid-like high-permittivity ones. A large set of results (not shown here) has confirmed that such a simple functional dependence on the invariants $T_1, T_2, T_4^E, T_5^E, T_6^E$ holds true irrespectively of the electromechanical properties of the matrix and particles, as measured by $\mu, \mu_p, \varepsilon, \varepsilon_p$, and irrespectively of the volume fraction of particles $c$. This functional behavior is admittedly remarkable. Indeed, the functional character of the macroscopic behavior of nonlinear heterogeneous material systems is in general markedly different from that of their constituents, but that is not the case here. Incidentally, this was already known to be the case for the overall nonlinear elastic response of Gaussian rubber isotropically filled with rigid particles (Lopez-Pamies et al., 2013a; Lopez-Pamies et al., 2013b), which corresponds to setting $\boldsymbol{E} = \mathbf{0}$ and $\mu_p = +\infty$ in the present context.

7.2 Solutions for isotropic distributions of spherical particles of poly- and mono-disperse sizes

The solution presented above corresponds to a two-phase isotropic particulate microstructure wherein the particles are infinitely polydisperse in size. In this section, we present solutions for the effective free-energy function $W$ of ideal elastic dielectric composites with two other classes of two-phase isotropic microstructures: $i)$ an isotropic distribution of polydisperse spherical particles of a finite number of different sizes and $ii)$ an isotropic distribution of monodisperse spherical particles. The motivation behind this choice stems from the fact that the vast majority of available experimental data corresponds to random isotropic distributions of filler particles that are roughly spherical in shape and of various sizes. Additionally, the analysis of these and the preceding iterative microstructures — ranging from infinitely polydisperse, to finitely polydisperse, to monodisperse — aims at shedding light on the effect of particle size dispersion on macroscopic properties.

The next two subsections describe the specifics of the two microstructures with spherical particles of interest here. The computation of their macroscopic elastic dielectric response is carried out by
means of hybrid finite elements, the technical details of which are deferred to Section 8.3. Section 7.2.3 presents the results for their macroscopic response in the limit of small deformations and moderate electric fields. The results for their macroscopic response under finite deformations and finite electric fields are presented in Section 7.2.4.

7.2.1 Spherical particles of polydisperse size

By definition, an isotropic distribution of spherical particles involves an infinite number of particles. Accounting for infinitely many particles is, of course, computationally not feasible. Here, we follow a well-established approximate approach and model isotropic distributions of polydisperse spherical particles as infinite media made out of the periodic repetition of a unit cell containing a random distribution of a large but finite number $N$ of spherical particles (see, e.g., Gusev, 1997; Michel et al., 1999; Segurado and LLorca, 2002; Lopez-Pamies et al., 2013b).

For convenience and without loss of generality, we select the defining unit cell to be a cube with edges of length $L = 1$. For definiteness, we consider that such a unit cell contains three families of spherical particles of distinct radii $R_p^{(i)}$ and volume fractions $c^{(i)}$ ($i = 1, 2, 3$) obeying the relations

\[
\{R_p^{(1)}, R_p^{(2)}, R_p^{(3)}\} = \left\{ R_p, \frac{7}{9} R_p, \frac{4}{9} R_p \right\} \quad \text{with} \quad R_p = L \left( \frac{3c^{(1)}}{4\pi N_p} \right)^{1/3} \quad (7.19)
\]

and

\[
\{c^{(1)}, c^{(2)}, c^{(3)}\} = \{0.5c, 0.25c, 0.25c\} \quad \text{with} \quad c^{(1)} + c^{(2)} + c^{(3)} = c, \quad (7.20)
\]

where $c$ is, again, the total volume fraction of particles in the composite and $N_p$ stands for the number of particles with the largest radius $R_p^{(1)} = R_p$ in the unit cell. Realizations within this class of microstructures are constructed with help of a random sequential adsorption algorithm (Lopez-Pamies et al., 2013b). Specifically, as a first step, $N_p$ particles of the largest radius $R_p^{(1)}$ are sequentially added to the unit cell until the condition $c^{(1)} = 0.5c$ is reached. Particles of the intermediate radius $R_p^{(2)}$ are added thereafter until the condition $c^{(1)} + c^{(2)} \approx 0.75c$ is satisfied. Particles with the smallest radius $R_p^{(3)}$ are then finally added until $c^{(1)} + c^{(2)} + c^{(3)} \approx c$. In general, this construction process yields microstructures that reach the target volume fraction $c$ only approximately (up to a small deviation that depends on the choice of the various parameters), thus the use of the symbol $\approx$ in the above expressions.
In order to construct realizations that allow for an adequate finite-element (FE) discretization, the random sequential adsorption algorithm that we employ enforces the following two constraints:

- The center-to-center distance between any two particles, $i$ and $j$ say with $i,j = 1, 2, ..., N$, must be greater than a certain minimum value $s_1$, adjusted by an offset factor $d_1 = 0.02$. This condition reads as

$$||X^i - X^j - h|| \geq s_1, \quad \text{with} \quad s_1 = (R_p^{(m_i)} + R_p^{(m_j)})(1 + d_1), \quad (7.21)$$

where $X^i$ ($X^j$) stands for the position of the center of particle $i$ ($j$), the superscripts $m_i, m_j = 1, 2, 3$ have been introduced to denote the sizes of the spheres $i$ and $j$, and $h$ is a vector with entries $0, L, -L$ for each of its three components in a Cartesian coordinate system aligned with the principal axes of the unit cell.

- The particles are not to be closer than a minimum distance $s_2$ to the boundaries of the unit cell, adjusted by an offset factor $d_2 = 0.05$. This condition reads as

$$|X^i_k - R_p^{(m_i)}| \geq s_2, \quad |X^i_k + R_p^{(m_i)} - L| \geq s_2, \quad s_2 = d_2 R_p^{(m_i)} \quad (k = 1, 2, 3), \quad (7.22)$$

for $i = 1, 2, \ldots, N$.

Guided by a parametric study aimed at identifying microstructures that (while not exactly) are practically isotropic, we utilize $N_p = 10$ which results into unit cells containing a total of $N = 36$ particles. Figure 7.4 illustrates two such unit cells generated by the algorithm described above for two volume fractions of particles: (a) $c = 0.05$ and (b) $c = 0.15$. To aid in the visualization of the entire microstructure, the figure also shows 27 contiguous unit cells out of the infinite medium.
considered. Particles with the smallest radius \( R_p^{(3)} \) are shown in blue, those with the intermediate radius \( R_p^{(2)} \) are shown in gray, and the particles with the largest radius \( R_p^{(1)} = R_p \) are shown in red.

### 7.2.2 Spherical particles of monodisperse size

Similar to the above approach for isotropic distributions of polydisperse spherical particles, isotropic distributions of monodisperse spherical particles are also modeled here as infinite media made out of the periodic repetition of a cubit unit cell containing a random distribution of a large but finite number \( N \) of spherical particles of the same size. It follows that the common radius of the particles is given by

\[
R_m = L \left( \frac{3c}{4\pi N} \right)^{1/3} \tag{7.23}
\]

in terms of the volume fraction \( c \) of particles in the composite and the total number of particles \( N \) in the unit cell. Realizations within this class of microstructures are generated by means of an adsorption algorithm that randomly and sequentially adds particles to the unit cell while enforcing the following two constraints (put in place, again, to allow for an adequate FE discretization):

\[
||X_i - X_j - \mathbf{h}|| \geq s_1, \quad \text{with} \quad s_1 = 2R_m(1 + d_1) \tag{7.24}
\]

and

\[
|X_k^i - R_m| \geq s_2, \quad |X_k^i + R_m - L| \geq s_2, \quad s_2 = d_2R_m \quad (k = 1, 2, 3) \tag{7.25}
\]

for \( i, j = 1, 2, \ldots, N \), where the offset factors are set to \( d_1 = 0.02 \) and \( d_2 = 0.05 \), as in the polydisperse case.

![Figure 7.5: Sample microstructures made out of the periodic repetition of a cubic unit cell with \( N = 30 \) randomly distributed spherical particles of identical size for two particle volume fractions: (a) \( c = 0.05 \) and (b) \( c = 0.15 \).](image)

A parametric study varying the number of particles indicates that, for our purposes, \( N = 30 \) particles are sufficient to achieve high degrees of isotropy. Figure 7.5 illustrates two unit cells with
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7.2.3 The limit of small deformations and moderate electric fields

Figure 7.6 shows FE solutions for the macroscopic elastic dielectric response in the limit of small deformations and moderate electric fields of the ideal elastic dielectric composites with the isotropic distribution of monodisperse spherical particles described above; see Chapter 5. In particular, plots are shown of the normalized values of the effective constants $\tilde{\mu}/\mu$, $\tilde{\varepsilon}/\varepsilon$, $\tilde{m}_K/\varepsilon$ — defined, again, by relations (7.8) with (7.9)–(7.10) — in terms of the volume fraction of particles $c$. Figures 7.6(a) and (b) display results for the case of stiff high-permittivity particles with $\mu_p = 10^2 \mu$ and $\varepsilon_p = 10^2 \varepsilon$, whereas Figs. 7.6(c) and (d) display results for liquid-like high-permittivity particles with $\mu_p = 10^{-1} \mu$ and $\varepsilon_p = 10 \varepsilon$. All the results correspond to the average of three realizations. In this regard, we note that the responses of all three realizations exhibited very small differences (less than 1%) between one another. Up to the volume fraction of particles considered $c = 0.25$, the corresponding solutions for the isotropic distribution of polydisperse spherical particles described above are virtually indistinguishable from those presented in Fig. 7.6 for monodisperse particles. To further shed light on this lack of sensitivity to size dispersion, Fig. 7.6 also includes the analytical result (dotted lines) given by (4.23)_{1,3,4} for an isotropic distribution of infinitely polydisperse spherical particles, the so-called differential coated sphere assemblage. It is evident that particle size dispersion has no effect whatsoever on the effective electromechanical constants $\tilde{\mu}$, $\tilde{\varepsilon}$, $\tilde{m}_K$ up to volume fractions of spherical particles of about $c = 0.2$. 

$N = 30$ particles of the same size for two different particle volume fractions: (a) $c = 0.05$ and (b) $c = 0.15$. The figure also shows 27 contiguous unit cells out of the infinite medium considered in order to help in the visualization of the entire microstructure.
Figure 7.6: Plots of the effective shear modulus $\tilde{\mu}/\mu$, effective permittivity $\tilde{\varepsilon}/\varepsilon$, and effective electrostrictive constant $\tilde{m}_{K}/\varepsilon$ of an ideal elastic dielectric, with initial shear modulus $\mu$ and initial permittivity $\varepsilon$, filled with an isotropic distribution of monodisperse spherical particles, with initial shear modulus $\mu_p$ and initial permittivity $\varepsilon_p$, all as functions of the volume fraction of particles $c$. Parts (a) and (b) show FE results (labeled “Sph. Mono. Exact” and displayed as solid circles) for stiff high-permittivity particles with $\mu_p = 10^2 \mu$ and $\varepsilon_p = 10^2 \varepsilon$. Parts (c) and (d) show FE results for liquid-like high-permittivity particles with $\mu_p = 10^{-1} \mu$ and $\varepsilon_p = 10 \varepsilon$. The corresponding analytical results (4.23) for the effective constants of an isotropic distribution of infinitely polydisperse spherical particles (labeled “Diff. Coat. Sph.” and displayed as dotted lines) are also included in the plots for comparison purposes.
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7.2.4 Finite deformations and finite electric fields

Figures 7.7 and 7.8 show FE solutions for the effective free-energy function (2.18) of the ideal elastic dielectric composites with the isotropic distribution of monodisperse spherical particles described in Section 7.2.2; again, the technical details of the FE calculations are presented in Section 8.3. Similar to the plots shown in Figs. 7.2 and 7.3 for the iterative microstructure, with the objective of gaining both qualitative and quantitative insight, Figs. 7.7 and 7.8 show plots of the normalized free energy $\tilde{W}/\mu$ as a function of each of the five normalized invariants $I_1, I_2, \varepsilon I_4^E/\mu, \varepsilon I_5^E/\mu, \varepsilon I_6^E/\mu$ for fixed values of the remaining four invariants. The results in Fig. 7.7 correspond to stiff high-permittivity particles with volume fraction $c = 0.05$ and material parameters $\mu_p = 10^2 \mu$ and $\varepsilon_p = 10^2 \varepsilon$, whereas those in Fig. 7.8 correspond to liquid-like high-permittivity particles with volume fraction $c = 0.15$ and material parameters $\mu_p = 10^{-1} \mu$ and $\varepsilon_p = 10 \varepsilon$. We recall that fixing the values of four of the invariants $I_1, I_2, \varepsilon I_4^E/\mu, \varepsilon I_5^E/\mu, \varepsilon I_6^E/\mu$ restricts the range of physical values that the remaining invariant can take on. The results shown in Figs. 7.7 and 7.8 span the entire range of physically allowable values for each case that is presented.

The corresponding results for the isotropic distribution of polydisperse spherical particles described in Section 7.2.1 are virtually indistinguishable from those presented in Figs. 7.7 and 7.8 for monodisperse particles, at least up to the volume fractions of particles considered here $c = 0.25$. Thus, the dispersion in size of spherical particles has little effect (sufficiently away from percolation, of course) on the macroscopic response of this class of ideal elastic dielectric composites, not only in the limit of small deformations and moderate electric fields as discussed in the context of Fig. 7.6, but more generally for finite deformations and finite electric fields.

The second and more important observation from the results shown in Figs. 7.7 and 7.8 is that the effective free-energy function (2.18) of ideal elastic dielectrics filled with spherical particles, much like that of the elastic dielectric composite with the iterative microstructure discussed in the preceding section, depends roughly linearly on the invariants $I_1, I_4^E, I_5^E$ and is roughly independent of $I_2, I_6^E$. For this class of microstructures too, the dependence on $I_4^E$ is much weaker than on $I_1$ and $I_5^E$. A broad range of results (in addition to those presented here) have confirmed that such a functional dependence on the invariants $I_1, I_2, I_4^E, I_5^E, I_6^E$ holds true irrespectively of the electromechanical properties of the matrix and particles, as measured by $\mu, \mu_p, \varepsilon, \varepsilon_p$, and irrespectively of the volume fraction of particles $c$. 


Figure 7.7: Plots of the effective free-energy function (2.18) of an ideal elastic dielectric, with initial shear modulus $\mu$ and initial permittivity $\varepsilon$, filled with an isotropic distribution of stiff high-permittivity monodisperse spherical particles, with initial shear modulus $\mu_p = 10^2 \mu$ and initial permittivity $\varepsilon_p = 10^2 \varepsilon$, at volume fraction $c = 0.05$. Results are shown for the values of the normalized free energy $W/\mu$ in terms of each of the five normalized invariants $\bar{T}_1, \bar{T}_2, \varepsilon\bar{T}_4/\mu, \varepsilon\bar{T}_5/\mu, \varepsilon\bar{T}_6/\mu$ for two sets of fixed values of the remaining four invariants. The solid lines (labeled “Sph. Mono. Exact”) correspond to the FE solution, while the dashed lines (labeled “Sph. Mono. Approx.”) correspond to the closed-form approximation (7.26).
Figure 7.8: Plots of the effective free-energy function (2.18) of an ideal elastic dielectric, with initial shear modulus $\mu$ and initial permittivity $\varepsilon$, filled with an isotropic distribution of liquid-like high-permittivity monodisperse spherical particles, with initial shear modulus $\mu_p = 10^{-1}\mu$ and initial permittivity $\varepsilon_p = 10\varepsilon$, at volume fraction $c = 0.15$. Results are shown for the values of the normalized free energy $W/\mu$ in terms of each of the five normalized invariants $I_1$, $I_2$, $\varepsilon T_4^E/\mu$, $\varepsilon T_5^E/\mu$, $\varepsilon T_6^E/\mu$ for two sets of fixed values of the remaining four invariants. The solid lines (labeled “Sph. Mono. Exact”) correspond to the FE solution, while the dashed lines (labeled “Sph. Mono. Approx.”) correspond to the closed-form approximation (7.26).
7.3 An approximate closed-form solution

The common functional dependence on the applied electromechanical loading exhibited by the three solutions presented above for different classes of isotropic particulate microstructures prompts the following approximate closed-form solution for the effective free-energy function of ideal elastic dielectric composites with any type of non-percolative isotropic particulate microstructure:

\[
\mathcal{W}(\mathbf{F}, \mathbf{E}, c) = \begin{cases} 
\frac{\bar{\mu}}{2} (\mathbf{T}_1 - 3) + \frac{\bar{m}_K - \bar{\varepsilon}}{2} \mathbf{T}_4 - \frac{\bar{m}_K}{2} \mathbf{T}_5 & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases} \tag{7.26}
\]

Here, we recall that \(\mathbf{T}_1, \mathbf{T}_4^E, \mathbf{T}_5^E\) stand for the standard \((\mathbf{F}, \mathbf{E})\)-based invariants defined by (7.3)\(_{1,3,4}\) and \(\bar{\mu}, \bar{\varepsilon}, \bar{m}_K\) denote the effective shear modulus, effective permittivity, and effective electrostrictive constant defined by (7.8) in the limit of small deformations and moderate electric fields. Again, for any given isotropic distribution of particles, the evaluation of these effective constants amounts to solving the two uncoupled linear pdes (7.9) and (7.10).

From a practical point of view, we remark that while the second-order linear elliptic pdes (10.20) and (10.21) for the tensorial fields \(\Gamma\) and \(\gamma\) cannot be directly solved in commercial FE codes, it is possible to make use of commercial FE codes to compute combinations of the components of their gradients that suffice to determine the effective constants \(\bar{\mu}, \bar{\varepsilon}, \bar{m}_K\) for any given microstructure. This is why and how. Because of the overall isotropy of the composites of interest here, as already discussed in Chapter 4, the effective coefficients (7.8) can be alternatively written as

\[
\begin{align*}
\bar{\mu} &= \int_{\Omega} 2\mu(\mathbf{X}) K_{12,s} \Gamma_{r12,s} d\mathbf{X}, \\
\bar{\varepsilon} &= \int_{\Omega} \frac{1}{3} \varepsilon(\mathbf{X}) \gamma_{m,m} d\mathbf{X}, \\
\bar{m}_K &= \int_{\Omega} 2\varepsilon(\mathbf{X}) \Gamma_{r12,s} K_{rspq} \gamma_{p,1} \gamma_{q,2} d\mathbf{X},
\end{align*} \tag{7.27}
\]

where it is recalled that \(K_{ijkl} = 1/2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - 1/3\delta_{ij}\delta_{kl}\). That is, knowledge of the components \(\Gamma_{r12,s}\) and \(\gamma_{i,j}\) of the gradients of the fields \(\Gamma\) and \(\gamma\) suffices for the computation of the effective constants \(\bar{\mu}, \bar{\varepsilon}, \bar{m}_K\). These components can be directly computed in any commercial FE code that can solve standard linear elastostatics and electrostatics problems. Indeed, it follows from (10.20)
that $\Gamma_{12,j}$ agrees identically with the gradient $u_{i,j}$ of the displacement field $u_i$ solution of the linear elastostatics problem

$$
\begin{cases}
\mu(X)(u_{i,j} + u_{j,i}) + \frac{1}{2}q_{ij} = 0 \\
u_{k,k} = 0
\end{cases}
$$

for $X \in \Omega$, $u_i = \delta_{i1}X_2$ for $X \in \partial\Omega$. \hfill (7.28)

It follows similarly from (10.21) that $\gamma_{i,j}$ agrees identically with the gradient $\Phi_j$ of the electric potential $\Phi$ solution of the linear electrostatics problem

$$
[\varepsilon(X)\Phi_j]_j = 0 \quad \text{for} \quad X \in \Omega, \quad \Phi = X_j \quad j = 1, 2, 3 \quad \text{for} \quad X \in \partial\Omega. \hfill (7.29)
$$

Having generated numerical solutions for $\Gamma_{r12,s}$ and $\gamma_{i,j}$, the integrals (7.27) can be readily evaluated by means of a quadrature rule to generate in turn the numerical solutions for the effective constants $\tilde{\mu}$, $\tilde{\varepsilon}$, and $\tilde{\mu}_K$.

The macroscopic constitutive relation (2.17) implied by the effective free-energy function (7.26) is given by

$$
\mathbf{S} = \tilde{\mu} \mathbf{F} + \tilde{\mu}_K \mathbf{F}^{-T} \mathbf{E} \otimes \mathbf{F}^{-T} \mathbf{F}^{-1} \mathbf{E} - \mathbf{p} \mathbf{F}^{-T}, \hfill (7.30)
$$

where $\mathbf{p}$ stands for the arbitrary hydrostatic pressure associated with the incompressibility constraint $\mathbf{J} = 1$, and

$$
\mathbf{D} = (\tilde{\varepsilon} - \tilde{\mu}_K) \mathbf{E} + \tilde{\mu}_K \mathbf{F}^{-1} \mathbf{F}^{-T} \mathbf{E}. \hfill (7.31)
$$

By construction, the effective free energy (7.26) is exact in the limit of small deformations and moderate electric fields as $\mathbf{F} \to \mathbf{I}$ and $\mathbf{E} \to \mathbf{0}$. Also by construction, while not exact, the effective free energy (7.26) is expected to be extremely accurate for finite deformations and finite electric fields given that it is linear in $\mathbf{T}_1$, $\mathbf{T}_4$, $\mathbf{T}_5^E$ and independent of $\mathbf{T}_2$, $\mathbf{T}_6^E$. This expectation is supported by the direct comparisons shown above in Figs. 7.2, 7.3, 7.7, 7.8 and further below in Figs. 7.10, 7.11 with the exact solutions for the three considered microstructures. Indeed, the results based on (7.26), shown as dashed lines in the figures, are seen to agree remarkably well, both qualitatively and quantitatively, with all exact results for all choices of matrix and particle material parameters $\mu$, $\mu_p$, $\varepsilon$, $\varepsilon_p$, as well as for all choices of volume fractions of particles $c$.

### 7.3.1 The $\mathbf{F}$ and $\mathbf{D}$ formulation

Depending on the specific problem at hand, it may be more convenient to utilize the macroscopic electric displacement field $\mathbf{D}$ as the independent electric variable instead of the macroscopic electric
field $\mathbf{E}$ utilized in (7.26). This can be readily accomplished with help of the following partial Legendre transform:

$$
\mathcal{W}^*(\mathbf{F}, \mathbf{D}, \mathbf{c}) = \sup_{\mathbf{E}} \left\{ \mathbf{D} \cdot \mathbf{E} + \mathcal{W}(\mathbf{F}, \mathbf{E}, \mathbf{c}) \right\}
$$

$$
= \left\{ \begin{array}{ll}
\frac{\mu}{2} [\mathcal{T}_1 - 3] + \\
\frac{1}{2\bar{m}_K} \left[ \mathcal{T}_5^D + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right)^2 \mathcal{T}_4^D + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) [\mathcal{T}_1 \mathcal{T}_5^D - \mathcal{T}_6^D] \right] \\
\infty
\end{array} \right.
$$

if $\mathcal{J} = 1$,

$$
\text{otherwise}
$$

(7.32)

where the invariants $\mathcal{T}_1, \mathcal{T}_2$ are given, again, by expressions (7.3)1,2, and

$$
\mathcal{T}_4^D = \mathbf{D} \cdot \mathbf{D}, \quad \mathcal{T}_5^D = \mathbf{F} \mathbf{D} \cdot \mathbf{F} \mathbf{D}, \quad \mathcal{T}_6^D = \mathbf{F}^T \mathbf{D} \cdot \mathbf{F}^T \mathbf{F} \mathbf{D}.
$$

(7.33)

Physically, the potential $\mathcal{W}^*$ defined by (7.32) corresponds to the macroscopic Helmholtz free energy of the composite. It follows that the first Piola-Kirchhoff stress tensor $\mathbf{S}$ and electric field $\mathbf{E}$ can be written in terms of the deformation gradient $\mathbf{F}$ and electric displacement $\mathbf{D}$ simply as

$$
\mathbf{S} = \frac{\partial \mathcal{W}^*}{\partial \mathbf{F}}(\mathbf{F}, \mathbf{D}, \mathbf{c}) - \eta \mathbf{F}^{-T}
$$

$$
= \mu \mathbf{F} - \eta \mathbf{F}^{-T} + \frac{1}{\bar{m}_K} \left[ \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) \mathcal{T}_5^D \mathbf{F} - \mathbf{F} \mathbf{D} \otimes \mathbf{F}^T \mathbf{F} \mathbf{D} - \mathbf{F} \mathbf{F}^T \mathbf{D} \otimes \mathbf{D} \right] + \left( 1 + \mathcal{T}_1 \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) \mathbf{F} \mathbf{D} \otimes \mathbf{D}
$$

$$
\frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \mathcal{T}_5^D + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right)^3 \mathcal{T}_4^D + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right)^2 [\mathcal{T}_1 \mathcal{T}_5^D - \mathcal{T}_6^D]
$$

$$
\frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \left[ \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) \mathcal{T}_5^D + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right)^2 \mathcal{T}_4^D + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) \mathcal{T}_6^D \right]
$$

$$
\left[ \mathbf{F} - \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) \mathbf{F}^{-T} \mathbf{F}^{-1} \mathbf{F}^{-T} \right],
$$

(7.34)
where \( \bar{q} \) stands for the arbitrary hydrostatic pressure associated with the incompressibility constraint \( \bar{J} = 1 \), and

\[
\bar{E} = \frac{\partial \bar{W}^*}{\partial \bar{D}}(\bar{F}, \bar{D}, c)
\]

\[
= \frac{1}{\bar{m}_K} \left[ 1 + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right)^3 + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right)^2 I_2 + \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) I_1 \right]
\]

\[
\left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right)^2 \bar{D} - \left( \frac{\bar{\varepsilon} - \bar{m}_K}{\bar{m}_K} \right) \bar{F}^T \bar{F} \bar{F}^T \bar{F} \bar{D}.
\]

(7.35)

We note that the (finite branch of the) effective free energy (7.32) in terms of \( \bar{F} \) and \( \bar{D} \) is not of the separable form \( \bar{W} = \bar{W}^{\text{elas}}(I_1, I_2, c) + \bar{W}^{\text{elec}}(I_4^D, I_5^D, I_6^D, c) \), which has been otherwise suggested in the literature based on grounds of simplicity (see, e.g., Ponte Castañeda and Siboni, 2012). This is in contrast to the form (7.26) of the effective free energy written in terms of \( \bar{E} \) and \( \bar{D} \), which is of the separable form \( \bar{W} = \bar{W}^{\text{elas}}(I_1, I_2, c) + \bar{W}^{\text{elec}}(I_4^E, I_5^E, I_6^E, c) \). We further note that the effective free energy (7.32) does depend on all five invariants \( I_1, I_2, I_4^D, I_5^D, I_6^D \), whereas (7.26) is independent of \( I_2 \) and \( I_6^E \).

### 7.3.2 Material instabilities

In addition to facilitating the computation of the macroscopic electromechanical constitutive response (7.34)–(7.35), the free-energy function (7.32) in terms of \( \bar{F} \) and \( \bar{D} \) provides the means to conveniently determine the possible onset of two classes of material instabilities: ii) instabilities associated with electromechanical limit loads and ii) microstructural instabilities of long wavelength. The former are characterized by the loss of positive definiteness of the tangent modulus of \( \bar{W}^* \) as defined by failure of the condition (see, e.g., Hill, 1957; Zhao and Suo, 2007)

\[
\min_{\bar{F}, \bar{D} \neq 0} \left\{ \bar{F}_{ij} \mathcal{E}_{ijkl}(\bar{F}, \bar{D}, c) \bar{F}_{kl} + 2 \bar{F}_{ij} \mathcal{M}_{ijkl}(\bar{F}, \bar{D}, c) \bar{D}_{kl} + \bar{D}_{ij} \mathcal{B}_{ijkl}(\bar{F}, \bar{D}, c) \bar{D}_{kl} \right\} > 0.
\]

(7.36)

On the other hand, long-wavelength instabilities are expected to be characterized by the loss of strong ellipticity of \( \bar{W}^* \), or, in other words, the loss of positive definiteness of its electromechanical acoustic tensor as defined by failure of the condition (see, e.g., Geymonat et al., 1993; Spinelli and Lopez-Pamies, 2015)

\[
\min_{\|u\| = \|v\| = 1, \ u, v \neq 0} \left\{ v \cdot \left[ \hat{K} - \frac{2}{(\text{tr}B)^2 - \text{tr}B^2} \hat{R} \left( \text{tr}(\hat{B})\hat{I} - \hat{B} \right) \hat{R}^T \right] v \right\} > 0.
\]

(7.37)
where $\hat{K} = i \hat{K} \hat{I}$, $\hat{R} = i \hat{R} \hat{I}$, $\hat{B} = i \hat{B} \hat{I}$ with

$$K_{ik} = F_{ja} F_{lb} \tilde{Z}_{lakh}(F, D, c) u_j u_l,$$

$$R_{ik} = F_{ja} F^{-1}_{bk} \tilde{M}_{lakh}(F, D, c) u_j,$$

$$B_{ij} = F_{ai} F^{-1}_{bj} \tilde{B}_{ab}(F, D, c),$$

$$\hat{I}_{ij} = \delta_{ij} - u_i u_j.$$

(7.38)

In the above expressions,

$$\tilde{Z}_{ijkl}(F, D, c) = \frac{\partial^2 W^*}{\partial F_{ij} \partial F_{kl}}(F, D, c),$$

$$\tilde{M}_{ijk}(F, D, c) = \frac{\partial^2 W^*}{\partial F_{ij} \partial D_k}(F, D, c),$$

$$\tilde{B}_{ij}(F, D, c) = \frac{\partial^2 W^*}{\partial D_i \partial D_j}(F, D, c),$$

(7.39)

where, with a slight abuse of notation, $W^*$ in these derivatives stands for the finite branch of the effective free energy (7.32).

The sets of all critical points $(F_{cr}, D_{cr})$ at which conditions (7.36) and (7.37) first fail along a continuous isochoric loading path, with starting point the ground state $(F, D) = (I, 0)$, define failure surfaces corresponding, respectively, to the attainment of electromechanical limit loads and the possible onset of long-wavelength instabilities. It is beyond the scope of this dissertation to study these failure surfaces in their entirety, but we do study the possible failure of conditions (7.36) and (7.37) for electromechanical boundary conditions encountered in typical electrostriction experiments, which are described next.

### 7.3.3 Electrostriction

Similar to Chapter 6, we focus here on one of the archetype experiments to probe the performance of dielectric elastomers that consists in exposing them to a uniaxial electric field and measuring the resulting deformation, commonly referred to as electrostriction. In practice, as shown schematically in Fig. 7.9, this is accomplished by sandwiching a thin layer of material between two compliant electrodes connected to a battery (see, e.g., Section 2.25 in Stratton, 1941; Pelrine et al., 1998; Di Lillo et al., 2011). In such a setup, the macroscopic stress is roughly zero everywhere (inside the material as well as in the surrounding space), while the macroscopic electric field is roughly uniform within the material and zero outside of it.
In this subsection, we study the specialization of the effective free-energy function (7.26) to the above-described boundary conditions of electrostriction. This seeks to shed light on whether the mere addition of (semi-)conducting/high-permittivity particles to dielectric elastomers — as modeled here thus far, without accounting for any other physical features such as the presence of interphases or viscous/dielectric dissipation — can indeed result in the drastic enhancement of electromechanical properties that has been observed experimentally (see, e.g., Zhang et al., 2002; Huang and Zhang, 2004; Huang et al., 2005; Carpi and De Rossi, 2005; Mc Carthy et al., 2009; Meddeb and Ounaies, 2012; Liu et al., 2013). It also seeks to shed light on the effect that the addition of particles has on electromechanical limit loads and on the onset of long-wavelength instabilities.

Macroscopic response. Consider hence macroscopic electromechanical states where the first Piola-Kirchhoff stress $\mathbf{S}$ and electric field $\mathbf{E}$ are of the form

$$\mathbf{S}_{ij} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \mathbf{E}_i = \begin{bmatrix} 0 \\ 0 \\ \Phi \end{bmatrix}. \quad (7.40)$$

Throughout this subsection, the components of any tensorial quantity are referred to the Cartesian laboratory axes $\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3$ depicted in Fig. 7.9. It follows from the constitutive relations (7.30) and (7.31) that

$$\mathbf{F}_{ij} = \begin{bmatrix} \lambda^{-1/2} & 0 & 0 \\ 0 & \lambda^{-1/2} & 0 \\ 0 & 0 & \lambda \end{bmatrix} \quad \text{and} \quad \mathbf{D}_i = \begin{bmatrix} 0 \\ 0 \\ \mathcal{D} \end{bmatrix}, \quad (7.41)$$
where the electrostriction stretch $\lambda$ in the direction of the applied electric field (see Fig. 7.9) and the non-trivial component $\mathbf{D}$ of the electric displacement are defined by the relations

$$\lambda^4 - \lambda + \frac{\tilde{m}_K}{\mu} \mathbf{E}^2 = 0 \quad \text{and} \quad \mathbf{D} = \left[ \bar{\varepsilon} - \tilde{m}_K \left( 1 - \frac{1}{\lambda^2} \right) \right] \mathbf{E}$$

(7.42)
in terms of the applied electric field $\mathbf{E}$.

In the absence of filler particles when $c = 0$, $\bar{\mu} = \mu$ and $\bar{\varepsilon} = \tilde{m}_K = \varepsilon$, and hence the constitutive relations (7.42) reduce of course to those of the unfilled elastic dielectric matrix: $\lambda^4 - \lambda + \varepsilon \mathbf{E}^2 / \mu = 0$ and $\mathbf{D} = \varepsilon \mathbf{E} / \lambda^2$. By comparing these to the general form of (7.42) for $c \neq 0$, it is plain that the addition of particles can potentially have a significant effect on the electrostriction response of the material depending on the ratio $\tilde{m}_K / \bar{\mu}$ and the values of $\bar{\varepsilon}$ and $\tilde{m}_K$. Sample numerical results are provided below in Figs. 7.10 and 7.11 for two cases of practical relevance.

**Material instabilities.** Along an electromechanical loading path of the form (7.40) where $| \mathbf{E} |$ is continuously increased from the ground state $\mathbf{E} = 0$, condition (7.36) first fails when

$$| \mathbf{E} | = \sqrt{\frac{3\bar{\mu}}{2^{8/3} \tilde{m}_K}} = \mathbf{E}_{LPD}. \quad (7.43)$$

The corresponding critical values $\lambda_{LPD}$ and $\mathbf{D}_{LPD}$ of the electrostriction stretch $\lambda$ and magnitude of the electric displacement $| \mathbf{D} |$ are given by

$$\lambda_{LPD} = 2^{-2/3} \quad \text{and} \quad \mathbf{D}_{LPD} = \left| \bar{\varepsilon} + \left( 2^{4/3} - 1 \right) \tilde{m}_K \right| \sqrt{\frac{3\bar{\mu}}{2^{8/3} \tilde{m}_K}}. \quad (7.44)$$

We can readily deduce from (7.43) that for microstructures and particle behaviors for which $\bar{\mu}/\tilde{m}_K < \mu/\varepsilon$, the addition of filler particles shifts the limiting electric field $\mathbf{E}_{LPD}$ to smaller values than that of the matrix $\sqrt{3\mu/2^{8/3} \varepsilon}$. By contrast, for microstructures and particle behaviors for which $\bar{\mu}/\tilde{m}_K > \mu/\varepsilon$, the addition of filler particles delays the attainment of the electromechanical limit load. Further, in view of (7.44)\textsubscript{1}, rather interestingly, the critical stretch at which the electromechanical limit load is attained is a constant and thus independent of the material parameters of the matrix and particles and also of the content of particles. The critical electric displacement (7.44)\textsubscript{2}, much like the critical electric field (7.43), does depend on the ratio $\bar{\mu}/\tilde{m}_K$ as well as on the values of $\bar{\varepsilon}$ and $\tilde{m}_K$, and hence it can be made to increase or decrease with the addition of particles depending on the specifics of the underlying microstructure and particle behavior. Sample numerical results for $\lambda_{LPD}$ and $\mathbf{E}_{LPD}$ are presented in Figs. 7.10 and 7.11.
As opposed to condition (7.36) and irrespectively of the material parameters of the matrix and particles and also of the content of particles, the strong ellipticity condition (7.37) never fails under conditions of electrostriction (7.40).

**Sample results.** Figures 7.10 and 7.11 present sample results determined from the proposed approximate effective free energy (7.26) — labeled “Aprox.” and displayed as dashed lines — for the macroscopic response and stability of an ideal elastic dielectric (with material parameters \( \mu \) and \( \varepsilon \)), filled with the infinitely polydisperse and monodisperse\(^2\) isotropic distributions of ideal elastic dielectric particles (with material parameters \( \mu_p, \varepsilon_p \), and volume fraction \( c \)) described in Sections 7.1 and 7.2, under the conditions of electrostriction (7.40). In particular, Fig. 7.10 presents results for stiff high-permittivity particles with material parameters \( \mu_p = 10^2 \) and \( \varepsilon_p = 10^2 \varepsilon \), whereas Fig. 7.11 presents results for liquid-like high-permittivity particles with material parameters \( \mu_p = 10^{-1} \) and \( \varepsilon_p = 10 \varepsilon \). Consistent with all preceding figures, the results pertaining to the iterative microstructure are labeled “HJ”, while those wherein the particles are monodisperse spheres are labeled “Sph. Mono.”. To further illustrate the accuracy of (7.26), the numerical viscosity solutions for the infinitely polydisperse iterative microstructure and the FE solutions for the microstructure with spherical particles are also included in the figures (up to the point at which we were able to compute them). These solutions are labeled as “Exact” and are displayed as solid lines or solid circles. The response of the unfilled elastic dielectric matrix (dotted line) is also displayed in the figures for comparison purposes.

Specifically, Figs. 7.10(a) through (d) show plots of the electrostriction stretch \( \lambda \) in terms of the applied electric field \( \vec{E} \), normalized by the quantity \( \sqrt{\mu/\varepsilon} \), as characterized by equation (7.42)\(_1\). Figures 7.10(a) and (c) correspond to the iterative microstructure at volume fractions of particles \( c = 0.05 \) and \( c = 0.15 \), respectively. On the other hand, Figs. 7.10(b) and (d) correspond to the microstructure with monodisperse spherical particles at the same volume fractions \( c = 0.05 \) and \( c = 0.15 \). An immediate observation from these four sets of plots is that the isotropic addition of stiff high-permittivity particles, irrespectively of the specifics of the underlying microstructure, has little effect on the electrostriction of ideal elastic dielectrics. Indeed, the composite with the iterative microstructure is seen to undergo a slightly smaller electrostriction

\(^2\)Again, for the volume fractions of particles considered here \( c \in [0,0.25] \), the results for isotropic distributions of spherical particles are insensitive to the dispersion in size of the particles. Hence, the results presented in Figs. 7.10 and 7.11 for monodisperse spherical particles can be viewed as corresponding to isotropic distributions of polydisperse spherical particles as well.
Figure 7.10: Electrostriction response and stability determined from the effective free energy (7.26) under conditions (7.40) — labeled “Approx.” and displayed as dashed lines in the plots — for an ideal elastic dielectric of initial shear modulus $\mu$ and initial permittivity $\varepsilon$ filled with stiff high-permittivity particles of initial shear modulus $\mu_p = 10^2 \mu$, initial permittivity $\varepsilon_p = 10^2 \varepsilon$, and volume fraction $c$. Results are shown for the infinitely polydisperse iterative microstructure (labeled “HJ”) and for the microstructure with monodisperse spherical particles (labeled “Sph. Mono.”). To further illustrate the accuracy of the closed-form approximation (7.26), corresponding plots are also included of the numerical viscosity solution for the iterative microstructure (labeled “Exact” and displayed as solid lines) and of the FE solutions for the microstructure with spherical particles (labeled “Exact” and displayed as solid lines or solid circles).
than the unfilled matrix for both volume fractions of particles considered, while the composite with spherical particles undergoes a slightly larger electrostriction than the unfilled matrix. These results for finite deformations and finite electric fields are in accord with the earlier findings reported in Section 6.1.2 in the asymptotic context of small deformations and moderate electric fields. By the same token, they are in disagreement with most experimental investigations, which have reported enhancements in electrostriction ranging from several tens (see, e.g., Liu et al., 2013) to several
thousands (see, e.g., Huang et al., 2005) of a percent for additions of stiff high-permittivity/(semi-)conducting particles at volume fractions $c < 0.1$. Thus, the results reported here provide evidence that the dominant microscopic mechanism by which the isotropic addition of stiff (semi-)conducting or high-permittivity particles leads to drastic enhancements in the electromechanical properties of dielectric elastomers is not the nonlinear elastic dielectric nature of these. We discuss this important result at greater length in Chapter 8.

Figures 7.10(a) through (d) also clearly illustrate the attainment of a maximum electric field at the critical stretch $\lambda_{LPD} = 2^{−2/3}$, independently of the specifics of the microstructure. The results beyond this critical stretch are shown for completeness. In practice, within the setup depicted in Fig. 7.9, electrostriction stretches $\lambda < \lambda_{LPD}$ would indeed be accessible if instead of a voltage, charges were applied on the compliant electrodes. The value of the maximum electric field $E_{LPD}$ given by relation (7.43), which does depend on the specifics of the microstructure, is plotted in Fig. 7.10(e) as a function of the volume fraction of particles $c$. Consistent with the previous remarks, the composite with the iterative microstructure exhibits modestly larger values of $E_{LPD}$ with the addition of particles. On the other hand, the composite with spherical particles exhibits modestly smaller values of $E_{LPD}$ as the content of particles is increased.

Figures 7.11(a) through (d) show results analogous to those shown in Figs. 7.10(a) through (e) for the case of liquid-like high-permittivity particles. As opposed to the addition of stiff high-permittivity particles, the addition of liquid-like high-permittivity particles is seen to have a significant effect on the electrostriction of ideal dielectrics. In particular, larger volume fractions of particles consistently lead to significantly larger electrostriction. These results for finite deformations and finite electric fields are also in accord with the earlier findings reported in Section 6.1.3 in the asymptotic context of small deformations and moderate electric fields. This is accompanied, however, by a significant reduction in the limiting electric field $E_{LPD}$. Finally, a quick glance at Fig. 7.11 suffices to recognize that both types of microstructures (“HJ” and “Sph. Mono.”) exhibit nearly identical behaviors. This suggests that the response of ideal elastic dielectrics isotropically filled with liquid-like high-permittivity particles is rather insensitive to fine microstructural details beyond the volume fraction of particles, more so than that of ideal elastic dielectrics filled with stiff high-permittivity particles.

In contrast to dielectric elastomers filled with stiff high-permittivity particles, there are comparatively few experimental investigations of dielectric elastomers filled with liquid-like high-permittivity particles (see, e.g., Fassler and Majidi, 2015). The theoretical results presented in Fig. 7.11 certainly
motivate more experimental studies of the latter.

**Beyond two-phase particulate dielectric elastomer composites.** The work presented so far in this document has focused on the fundamental idealization of dielectric elastomer composites as *two-phase* particulate composites. For any type of filled elastomer, however, it is well known that the “anchoring” of the underlying polymer chains to the filler particles forces the chains into conformations that are very different from those in the bulk, hence resulting in “interphases” of possibly several tens of nanometers in thickness and of different mechanical and physical behavior (see, e.g., Leblanc, 2010; Goudarzi et al., 2015 and references therein). In addition, some applications may favor the use of dielectric elastomers filled with particles of different materials (see, e.g., Dang et al., 2003; Nan et al., 2003). Accounting for these two features requires a microscopic description that views dielectric elastomer composites as *N-phase* particulate composites.

We conjecture that the proposed closed-form solution (7.26) provides an accurate approximation for the effective free-energy function of ideal elastic dielectric composites not just with any type of (non-percolative) isotropic two-phase particulate microstructure, but, more generally, with any type of (non-percolative) isotropic *N-phase* particulate microstructure:

$$
\overline{W}(\mathbf{F}, \mathbf{E}, c_p, c_i) = \min_{\mathbf{F} \in \mathcal{K}} \max_{\mathbf{E} \in \mathcal{E}} \int_{\Omega} W(\mathbf{X}, \mathbf{F}, \mathbf{E}) \, d\mathbf{X},
$$

(7.45)

where

$$
W(\mathbf{X}, \mathbf{F}, \mathbf{E}) = \begin{cases} 
\mu(\mathbf{X}) \left( 2[I_1 - 3] - \frac{\varepsilon(\mathbf{X})}{2} I_5^E \right) & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases}
$$

(7.46)

with

$$
\mu(\mathbf{X}) = [1 - \theta_p(\mathbf{X}) - \theta_i(\mathbf{X})] \mu + \theta_p(\mathbf{X}) \mu_p(\mathbf{X}) + \theta_i(\mathbf{X}) \mu_i(\mathbf{X})
$$

(7.47)

and

$$
\varepsilon(\mathbf{X}) = [1 - \theta_p(\mathbf{X}) - \theta_i(\mathbf{X})] \varepsilon + \theta_p(\mathbf{X}) \varepsilon_p(\mathbf{X}) + \theta_i(\mathbf{X}) \varepsilon_i(\mathbf{X}).
$$

(7.48)

Here, \( \theta_p \) and \( \theta_i \) denote the indicator functions of the spatial regions occupied in the ground state \( \Omega \) by the particles and by the surrounding interphases with *possibly pointwise heterogeneous* material parameters \( \mu_p = \mu_p(\mathbf{X}) \), \( \varepsilon_p = \varepsilon_p(\mathbf{X}) \) and \( \mu_i = \mu_i(\mathbf{X}) \), \( \varepsilon_i = \varepsilon_i(\mathbf{X}) \), respectively. This conjecture is based on the prevailing observation that the effective free energies \( \overline{W} \) of all of the isotropic composite materials with *pointwise* ideal elastic dielectric behavior that we have studied feature linearity in the invariants \( \mathcal{T}_1, \mathcal{T}_4^E, \mathcal{T}_5^E \) and are independent of \( \mathcal{T}_2, \mathcal{T}_6^E \), irrespectively of the specifics of their heterogeneity.
7. Nonlinear electroelastic deformations of DECs: Ideal elastic dielectrics

By way of an example, in support of the above conjecture, Fig. 7.12(a) illustrates the close agreement between the result (dashed line) determined from the approximation (7.26) and from a FE solution (solid line) for the $\lambda$ vs. $\bar{E}/\sqrt{\mu/\varepsilon}$ electrostriction response of a three-phase composite made out of an ideal elastic dielectric matrix, with material parameters $\mu$ and $\varepsilon$, filled with an isotropic distribution of monodisperse prolate spheroidal particles that are bonded to the matrix through interphases of the same constant thickness. The volume fraction of particles and their aspect ratio are $c_p = 0.035$ and $\omega = 1.5$, while the volume fraction of interphases is $c_i = 0.035$. The particles and interphases are homogeneous ideal elastic dielectrics with material parameters $\mu_p = 50\mu$, $\varepsilon_p = 50\varepsilon$ and $\mu_i = 5\mu$, $\varepsilon_i = 5\varepsilon$. The resulting effective electromechanical constants (7.8) are given by $\tilde{\mu} = 1.1583\mu$, $\tilde{\varepsilon} = 1.2044\varepsilon$, $\tilde{m}_K = 1.1783\varepsilon$. Figure 7.12(b) illustrates the unit cell whose periodic repetition defines the precise microstructure in this example.

![Figure 7.12](image_url)

Figure 7.12: (a) Comparison of the electrostriction response (see Section 7.3.3 for the definition of the pertinent variables) determined from the approximate closed-form free energy (7.26) and from a FE solution (labeled “Exact” and displayed as a solid line) of the three-phase isotropic ideal elastic dielectric composite whose defining unit cell is depicted in (b), namely, an ideal elastic dielectric matrix (with initial shear modulus $\mu$ and initial permittivity $\varepsilon$) filled with an isotropic distribution of homogeneous monodisperse prolate spheroidal particles that are bonded to the matrix through homogeneous interphases of the same constant thickness. The volume fraction $c_p$, aspect ratio $\omega$, and material parameters $\mu_p$, $\varepsilon_p$ of the particles are indicated in the figure, as so are the volume fraction $c_i$ and material parameters $\mu_i$, $\varepsilon_i$ of the interphases.
Nonlinear electroelastic deformations of dielectric elastomer composites: non-Gaussian elastic dielectrics

Naviguer est une activité qui ne convient pas aux imposteurs. Dans bien des professions, on peut faire illusion et bluffer en toute impunité. En bateau, on sait ou on ne sait pas.

– Éric Tabarly, Mémoires du large, 1998

In the preceding chapter, we determined a general homogenization solution for the macroscopic elastic dielectric response of Gaussian dielectric elastomers filled with any type of non-percolative isotropic distribution of ideal elastic dielectric particles. In this chapter, we make use of this fundamental result as a building block to construct in turn a general approximate analytical solution for the macroscopic response of isotropic dielectric elastomer composites with non-ideal elastic dielectric constituents. This is accomplished by means of a nonlinear comparison medium method. Complementary to this analytical approach, we also present a hybrid finite-element (FE) formulation to construct homogenization solutions numerically for the macroscopic elastic dielectric response of isotropic dielectric elastomer composites. Finally, we confront the above theoretical constitutive results to experimental data.
Non-Gaussian dielectric elastomers. Specifically, we consider in this chapter elastomeric matrix materials characterized by free-energy functions of the form

\[
W^{(1)}(F, E) = \begin{cases} 
\Psi(I_1) - \frac{\varepsilon}{2} I_5^E & \text{if } \det F = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]

(8.1)

where \(I_1 = F \cdot F, I_5^E = F^{-T} E \cdot F^{-T} E\), \(\Psi\) is any non-negative function of choice (suitably well-behaved) satisfying the linearization conditions

\[
\Psi(3) = 0 \text{ and } \Psi'(3) = \frac{\mu}{2}
\]

(8.2)

with \(\mu\) and \(\varepsilon\) denoting the initial shear modulus and the initial permittivity of the material; throughout this chapter, we make use of the convention \(y'(x) = \frac{dy(x)}{dx}\) to denote the derivative of functions of a single scalar variable. We recall that basic physical considerations dictate that \(\mu > 0\) and \(\varepsilon \geq \varepsilon_0\), where \(\varepsilon_0 \approx 8.85 \times 10^{-12} \text{ F/m}\) stands for the permittivity of vacuum.

We remark that free-energy functions of the form (8.1) have been shown to describe reasonably well the response of a broad variety of dielectric elastomers over wide ranges of deformations and electric fields (see, e.g., Wissler, 2007; Lopez-Pamies, 2010). While analytical results will be worked out in Section 8.2 for arbitrary choices of the function \(\Psi\), in Section 8.4 sample numerical results will be presented in particular for the choice

\[
\Psi(I_1) = \frac{3^{1-\alpha_1}}{2\alpha_1} \mu_1 [I_1^{\alpha_1} - 3^{\alpha_1}] + \frac{3^{1-\alpha_2}}{2\alpha_2} \mu_2 [I_1^{\alpha_2} - 3^{\alpha_2}].
\]

(8.3)

In this expression, \(\mu_1, \mu_2, \alpha_1, \alpha_2\) are real-valued material parameters that may be associated with the non-Gaussian statistical distribution of the underlying polymer chains. In addition to its mathematical simplicity and physical meaning of its parameters, we choose this class of functions because of its rich functional form and demonstrated descriptive and predictive capabilities (Lopez-Pamies, 2010).

For later use, we note that the partial Legendre transform (A.1) corresponding to the local free-energy function (8.1) is simply given by

\[
W^{(1)*}(F, D) = \sup_E \left\{D \cdot E + W^{(1)}(F, E)\right\} = \begin{cases} 
\Psi(I_1) + \frac{1}{2\varepsilon} I_5^D & \text{if } \det F = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]

(8.4)

where \(I_5^D = FD \cdot FD\).
Nonlinear elastic dielectric particles. On the other hand, we consider the elastic dielectric behavior of the filler particles to be characterized by free-energy functions of the form

\[
W^{(2)}(F, E) = \begin{cases} 
\frac{\mu_p}{2} [I_1 - 3] - S(I_5^E) & \text{if } \det F = 1, \\
+\infty & \text{otherwise}
\end{cases},
\]

(8.5)

where, again, \(I_1 = F \cdot F\), \(I_5^E = F^{-T} E \cdot F^{-T} E\), \(\mu_p\) stands for the initial shear modulus of the particles, and \(S\) is any function of choice satisfying the linearization conditions

\[
S(0) = 0 \quad \text{and} \quad S'(0) = \frac{\varepsilon_p}{2},
\]

(8.6)

with \(\varepsilon_p\) denoting the initial permittivity of the particles, and the convexity conditions

\[
S'(I_5^E) > 0 \quad \text{and} \quad S'(I_5^E) + 2I_5^E S''(I_5^E) > 0.
\]

(8.7)

The inequalities (8.7) on the function \(S\) ensure that the free energy (8.5) is, much like (8.1), concave in \(E\).

Free-energy functions of the form (8.5) are general enough to adequately model the elastic dielectric responses over wide ranges of deformations and electric fields of hard polymers such as polyaniline (PANI) and copper phthalocyanine oligomers (O-CuPc), ceramics such as titania (TiO\(_2\)), and liquids such as Galinstan, which have been utilized/suggested as filler particles in experimental investigations (see, e.g., Li et al., 2004; Huang et al., 2005; Liu et al., 2013; Fassler and Majidi, 2015). We emphasize in particular that free-energy functions of the form (8.5) are general enough to model (albeit ignoring dissipative effects) polarization saturation phenomena typical, for instance, of ferroelectric ceramics at large electric fields. In this case, noting that the polarization \(p\) (per unit deformed volume) implied by (8.5) is given by the expression

\[
p = -F \frac{\partial W_p}{\partial E}(F, E) - \varepsilon_0 F^{-T} E = [2S'(I_5^E) - \varepsilon_0] F^{-T} E,
\]

(8.8)

in addition to the linearization and convexity conditions (8.6)–(8.7), it must be required that

\[
S'(I_5^E) = \frac{\varepsilon_0}{2} + \frac{p_s}{2 \sqrt{I_5^E}} + o \left( \frac{1}{\sqrt{I_5^E}} \right)
\]

(8.9)

in the limit as \(I_5^E \to \infty\). In this last expression, the positive material constant \(p_s\) characterizes the magnitude of the saturated polarization. While analytical results will be worked out in Section 8.2
for any function \( S \) of choice, in Section 8.4 sample numerical results will be presented in particular for the classical polarization-saturation model due to Debye (Langevin, 1905; Debye, 1929):

\[
S(I^E_5) = \varepsilon_0 \frac{I^E_5}{2} + \frac{p_s^2}{3(\varepsilon_p - \varepsilon_0)} \left[ \ln \left( \sinh \left( \frac{3(\varepsilon_p - \varepsilon_0) \sqrt{I^F_5}}{p_s} \right) \right) - \ln \left( \frac{3(\varepsilon_p - \varepsilon_0) \sqrt{I^F_5}}{p_s} \right) \right].
\]

(8.10)

Also for later use, we note that the partial Legendre transform (A.1) corresponding to the local free-energy function (8.5) is given by

\[
W^{(2)}(F, D) = \sup_E \left\{ D \cdot E + W^{(2)}(F, E) \right\} = \begin{cases} \frac{\mu_p}{2} |I_1 - 3| + S^*(I^D_5) & \text{if } \det F = 1 \\ +\infty & \text{otherwise} \end{cases},
\]

(8.11)

where, again, \( I^D_5 = FD \cdot FD \) and \( S^*(I^D_5) = I^D_5 S'(z^{-1}(I^D_5))/2 - S(z^{-1}(I^D_5)) \) with \( z^{-1} \) denoting the inverse of the function \( z \) defined by \( z(x) = 4x(S'(x))^2 \). Given the properties (8.6)–(8.7) of the function \( S \), it follows that

\[
S^*(0) = 0, \quad S^{**}(0) = \frac{1}{2\varepsilon_p},
\]

(8.12)

and

\[
S^{**}(I^D_5) > 0, \quad S^{**}(I^D_5) + 2I^D_5 S^{***}(I^D_5) > 0,
\]

(8.13)

the latter of which implies that the free-energy function (8.11) is convex in \( D \), in agreement with its very definition.

**The macroscopic response.** Owing to the assumed isotropy of the microstructure and the constitutive isotropy and incompressibility of the matrix material (8.1) and filler particles (8.5), the resulting macroscopic elastic dielectric response is isotropic and incompressible. This implies that the effective free-energy function (2.18) in this case only depends on the macroscopic deformation gradient \( F \) and macroscopic Lagrangian electric field \( E \) through five invariants and becomes unbounded for non-isochoric deformations when \( \det F \neq 1 \). Consistent with the variables employed in Chapter 7, with a slight abuse of notation, we write

\[
\overline{W}(F, E, c) = \begin{cases} W(T_1, T_2, T^E_4, T^E_5, T^E_6, c) & \text{if } \det F = 1 \\ +\infty & \text{otherwise} \end{cases}
\]

(8.14)

in terms of the five standard invariants

\[
T_1 = F \cdot F, \quad T_2 = F^{-T} \cdot F^{-T}, \quad T^E_4 = E \cdot E,
\]

\[
T^E_5 = F^{-T} E \cdot F^{-T} E, \quad T^E_6 = F^{-1} F^{-T} E \cdot F^{-1} F^{-T} E.
\]

(8.15)
8. Nonlinear electroelastic deformations of DECs: Non-Gaussian elastic dielectrics

Similarly, we write the partial Legendre transform (A.3) of the effective free-energy function (8.14) as

$$\mathbb{W}^*(F, D, c) = \begin{cases} 
\mathbb{W}^*(I_1, I_2, I_4^D, I_5^D, I_6^D, c) & \text{if } \det F = 1 \\
+\infty & \text{otherwise}
\end{cases} \quad (8.16)$$

in terms of the five standard invariants (8.15) and

$$I_4^D = D \cdot D, \quad I_5^D = FD \cdot FD, \quad I_6^D = F^T FD \cdot F^T FD. \quad (8.17)$$

8.1 A nonlinear comparison medium method in nonlinear electroelastostatics

In this section, we put forth a comparison medium method that allows us to generate variational approximations for the effective free-energy function $\mathbb{W}$ of a given two-phase elastic dielectric composite, as defined by the variational problem (2.18), in terms of the effective free-energy function, $\mathbb{W}_0$ say, of another elastic dielectric composite (possibly comprising finitely or infinitely many phases). For clarity of exposition, the method is presented in its general form. Its specialization to the non-Gaussian dielectric elastomers isotropically filled with nonlinear elastic dielectric particles of interest in this chapter will be presented in Section 8.2.

We recall that comparison medium methods are analytical techniques that allow one to construct variational approximations for the macroscopic properties of a given composite material in terms of the macroscopic properties of another material. The latter is referred to as the comparison medium. The central defining idea of these methods can be traced back to the pioneering work of Talbot and Willis (1985) who laid out a complete comparison medium formalism — where the comparison medium is of arbitrary choice, possibly nonlinear and heterogeneous — for properties characterized by convex potentials of a single field. By introducing suitable partial Legendre transforms, Lopez-Pamies et al. (2013b) extended the general formalism of Talbot and Willis (1985) to nonlinear properties characterized by the class of non-convex potentials that are inherent to nonlinear elasticity. In this section, we work out an extension of the comparison medium method of Lopez-Pamies et al. (2013b) to the coupled realm of nonlinear electroelastostatics, where the properties of interest are characterized by non-convex potentials that are functions of two (one mechanical and one electrical) fields.
As will become apparent below, contrary to the minimax nature of the variational problem (2.18) defining \( \overline{W} \), the overall minimum nature of the variational problem (A.5) defining \( \overline{W}^* \) shall prove gainful in our strategy. We thus begin by considering an elastic dielectric composite characterized locally by the (possibly compressible and anisotropic) two-phase Helmholtz free-energy function \( W^*(X, F, D) = [1 - \theta(X)]W^{(1)*}(F, D) + \theta(X)W^{(2)*}(F, D) \), where the indicator function \( \theta \) is the same as in (2.13). Following Lopez-Pamies et al. (2013b), we consider as well an arbitrary comparison medium with pointwise free energy \( W_0^*(X, F, D) \), introduce the functions \( f = f(X, F, J, D) \) and \( f_0 = f_0(X, F, J, D) \) such that
\[
f(X, F, J, D) = W^*(X, F, D) = [1 - \theta(X)]W^{(1)*}(F, D) + \theta(X)W^{(2)*}(F, D) \quad \text{when} \quad J = \det F
\]  
and
\[
f_0(X, F, J, D) = W_0^*(X, F, D) \quad \text{when} \quad J = \det F,
\]
and define the Legendre transform
\[
(f - f_0)^*(X, P, Q, R) \doteq \sup_{F, J, D} [F \cdot P + JQ + D \cdot R - f(X, F, J, D) + f_0(X, F, J, D)].
\]  
Now, for any \( P, Q, \) and \( R \) it follows from (8.20) that
\[
f(X, F, J, D) \geq f_0(X, F, J, D) + F \cdot P + JQ + D \cdot R - (f - f_0)^*(X, P, Q, R)
\]
and therefore that
\[
\overline{W}^*(F, D, c) \geq \min_{F \in \mathcal{K}} \min_{D \in \mathcal{D}} \int_\Omega [f_0(X, F, J, D) + F \cdot P + JQ + D \cdot R] dX - \int_\Omega (f - f_0)^*(X, P, Q, R) dX,
\]  
where \( J = \det F \) here and henceforth. We recall that the minima sought in the right-hand side of (8.22) are over the sets \( \mathcal{K} \) and \( \mathcal{D} \) of admissible deformation gradients \( F(X) \) with prescribed volume average \( F \) and of admissible divergence-free electric displacement fields \( D(X) \) with volume average \( D \). A well-known property of minima of sums yields the further inequality
\[
\overline{W}^*(F, D, c) \geq \overline{W}_0^*(F, D) + \min_{F \in \mathcal{K}} \int_\Omega F \cdot P dX + \min_{F \in \mathcal{K}} \int_\Omega J Q dX + \min_{D \in \mathcal{D}} \int_\Omega D \cdot R dX - \int_\Omega (f - f_0)^*(X, P, Q, R) dX,
\]
where the notation \( \overline{W}_0^* \) has been introduced to denote the effective free-energy function of the comparison medium with local free energy \( f_0(X, F, J, D) = W_0^*(X, F, D) \):
\[
\overline{W}_0^*(F, D) = \min_{F \in \mathcal{K}} \min_{D \in \mathcal{D}} \int_\Omega f_0(X, F, J, D) dX.
\]
The second, third, and fourth terms in the right-hand side of (8.23) are bounded from below provided that $P$ is a divergence-free field, $Q$ is a constant, and $R$ is a curl-free field. For simplicity, we select all these fields to be constant and denote them by $P = P$, $Q = Q$, and $R = R$. After a standard calculation, this simplifying prescription gives

$$W^* (F, D, c) \geq W^*_0 (F, D) + \int_\Omega (f - f_0)^* (X, P, Q, R) \, dX,$$

(8.25)

where the definition $J = \det F$ has been employed.

The inequality (8.25) is valid for any choice of constant fields $P$, $Q$, and $R$, as well as for any choice of local free-energy function $f_0(X, F, J, D)$ describing the microstructure and local elastic dielectric behavior of the comparison medium. Optimizing with respect to $P$, $Q$, and $R$ yields

$$W^* (F, D, c) \geq W^*_0 (F, D) + \sup_{P, Q, R} \left\{ F \cdot P + J Q + D \cdot R - \int_\Omega (f - f_0)^* (X, P, Q, R) \, dX \right\},$$

(8.26)

Optimizing in turn with respect to $f_0$ leads formally to

$$W^* (F, D, c) \geq \sup_{f_0} \left\{ W^*_0 (F, D) + \left( \int_\Omega (f - f_0)^* dX \right)^* (F, J, D) \right\}. $$

(8.27)

### 8.1.1 A partially optimized explicit formulation

The computation of the optimized bound (8.27) involves two technical difficulties. The first one is that the function $(f - f_0)^*$ may exhibit corners, and hence the computation of the Legendre transform of its average in (8.27) may require the use of subgradients as opposed to standard differentiation; this is a difficulty that already appears in the classical context of convex energies of a single field (see, e.g., Willis, 1991). The second and more severe technical obstacle is that the supremum operation in (8.27) involves the optimization with respect to the microstructure of the comparison medium (in addition, as well, to the optimization with respect to its constitutive elastic dielectric behavior).

Such an optimization requires the computation of complicated integrals involving the product of different indicator functions — the indicator function $\theta(X)$ associated with $f$ and the indicator function, $\theta_0(X)$ say, associated with $f_0$ — in the second term of the right-hand side of (8.27). In this chapter, in the interest of analytical tractability, we shall be content with employing a partially optimized version of the result (8.25) — and not the fully optimized bound (8.27) — which avoids the two above-mentioned technical difficulties altogether.
Thus, in order to avoid the use of subgradients we set $\mathbf{F} = \mathbf{0}$, $\overline{\mathbf{Q}} = \mathbf{0}$, and $\mathbf{R} = \mathbf{0}$ so that, upon recognizing the string of equalities

$$(f-f_0)^*(X,0,0,0) = \sup_{A,a,B} [-f(X,A,a,B)+f_0(X,A,a,B)] = -\inf_{A,a,B} [f(X,A,a,B)-f_0(X,A,a,B)],$$

relation (8.25) reduces to

$$W^*(\mathbf{F},\mathbf{D},c) \geq W^*_0(\mathbf{F},\mathbf{D}) + \int_\Omega \inf_{A,a,B} [f(X,A,a,B)-f_0(X,A,a,B)] \, d\mathbf{X}. \quad (8.29)$$

Further, in order to avoid the computation of complicated integrals in (8.29) we restrict attention to comparison media with the same microstructure as the actual elastic dielectric composite, namely,

$$f_0(X,F,J,D) = [1-\theta(X)]f_0(F,J,D) + \theta(X)f_0(F,J,D), \quad (8.30)$$

where the indicator function $\theta$ is the same as in (2.13). With this choice, relation (8.29) leads$^1$ to the following lower bound for $\overline{W}^*$:

$$W^*(\mathbf{F},\mathbf{D},c) \geq W^*_0(\mathbf{F},\mathbf{D})+(1-c) \inf_{A,a,B} [f_\mathbf{\infty}(A,a,B)-f_0(A,a,B)]+c \inf_{A,a,B} [f_\mathbf{\infty}(A,a,B)-f_0(A,a,B)],$$

where, in analogy with (8.18), we have made use of the notation $f_\mathbf{\infty}(F,J,D) = W^{(1)*}(\mathbf{F},\mathbf{D})$ and $f_\mathbf{\infty}(F,J,D) = W^{(2)*}(\mathbf{F},\mathbf{D})$. Moreover, the symmetry of relation (8.29) in the pairs $(\overline{W}^*,f)$ and $(\overline{W}^*_0,f_0)$ implies as well the following upper bound:

$$W^*(\mathbf{F},\mathbf{D},c) \leq W^*_0(\mathbf{F},\mathbf{D})+(1-c) \sup_{A,a,B} [f_\mathbf{\infty}(A,a,B)-f_0(A,a,B)]+c \sup_{A,a,B} [f_\mathbf{\infty}(A,a,B)-f_0(A,a,B)]. \quad (8.32)$$

Note that the lower bound (8.31) is non-trivial provided that $f_\mathbf{\infty}-f_0 > -\infty$ and $f_\mathbf{\infty}-f_0 > -\infty$, while the upper bound (8.32) is non-trivial provided that $f_\mathbf{\infty}-f_0 < \infty$ and $f_\mathbf{\infty}-f_0 < \infty$. Combining the inequalities (8.31) and (8.32) leads to the following approximate solution for the effective free-energy

---

$^1$An alternative direct derivation of the formula (8.31) follows from a straightforward extension of the derivation of Willis (see, e.g., equation (3.3) in Willis, 1991; see also deBotton and Shmuel, 2010) of Ponte Castañeda’s bound (1991) in the context of convex energies of a single field: $\overline{W}^* = \min_{F \in K} \min_{D \in D} \int_\Omega [W_0^* + (W^* - W_0^*)] \, d\mathbf{X} = \overline{W}_0 + \int_\Omega \min(W^* - W_0^*) \, d\mathbf{X}$. 

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function (A.5):

\[
W^*(F, D, c) = \begin{cases} 
W^*_0(F, D) + (1-c) \min_{A,a,B} [f_n(A, a, B) - f_{0_n}(A, a, B)] + c \min_{A,a,B} [f_p(A, a, B) - f_{0_p}(A, a, B)] & \text{if } f_n - f_{0_n} > -\infty \\
\phantom{W^*_0(F, D) + (1-c) \min_{A,a,B} [f_n(A, a, B) - f_{0_n}(A, a, B)] + c \min_{A,a,B} [f_p(A, a, B) - f_{0_p}(A, a, B)]} & \text{if } f_p - f_{0_p} > -\infty \\
\end{cases}
\]

(8.33)

where the equality in this last expression has been used in the sense of a variational approximation.

The result (8.33) is valid for any choice of free-energy functions \( f_0_m \) and \( f_0_p \) describing the elastic dielectric behaviors of the matrix and filler particles in the comparison medium. Optimizing (8.33) with respect to these free energies leads formally to

\[
W^*(F, D, c) = \begin{cases} 
\sup_{f_0_m, f_0_p} \{ W^*_0(F, D) + (1-c) \min_{A,a,B} [f_n(A, a, B) - f_{0_n}(A, a, B)] + c \min_{A,a,B} [f_p(A, a, B) - f_{0_p}(A, a, B)] \} & \text{if } f_n - f_{0_n} < \infty \\
\phantom{\sup_{f_0_m, f_0_p} \{ W^*_0(F, D) + (1-c) \min_{A,a,B} [f_n(A, a, B) - f_{0_n}(A, a, B)] + c \min_{A,a,B} [f_p(A, a, B) - f_{0_p}(A, a, B)] \}} & \text{if } f_p - f_{0_p} < \infty \\
\end{cases}
\]

(8.34)

Expression (8.34) constitutes the main result of this section: it provides a variational approximation for the effective free-energy function (A.5) of a two-phase elastic dielectric composite with local free energy (8.18) in terms of the effective free-energy function (8.24) of another two-phase elastic dielectric composite with local free energy (8.30). For the case when the effective Helmholtz free-energy function \( W^* \) resulting from (8.34) is convex in \( D \), the corresponding effective free-energy function \( \overline{W} \) in terms of \( \overline{F} \) and \( \overline{E} \) can be expediently computed via the following partial Legendre transform:

\[
\overline{W}(F, E, c) = -\sup_\overline{D} \{ \overline{D} \cdot E - W^*(F, D, c) \}.
\]

(8.35)

It is plain that the usefulness of the variational approximation (8.34) — or more generally (8.33) — hinges upon having knowledge of the effective free-energy function \( \overline{W}^*_0 \) for the comparison medium.

While there have been no prior results available heretofore, we now have at our disposal the results for ideal elastic dielectric composites worked out in Chapter 7.
8.2 Application to non-Gaussian dielectric elastomers

isotropically filled with nonlinear elastic dielectric particles

In the sequel, we make use of the ideal elastic dielectric composites considered in Chapter 7 as choices for the comparison medium in the formulation (8.34)–(8.35) in order to construct an approximate homogenization solution for the effective free energy (2.18) of non-Gaussian dielectric elastomers, characterized by free-energy functions of the form (8.1), isotropically filled with nonlinear elastic dielectric particles characterized by free-energy functions of the form (8.5).

We begin by setting

\[ f_m(F, J, D) = \begin{cases} \Psi(I_1) + \frac{1}{2\varepsilon_m} I_5^D & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \]

\[ f_p(F, J, D) = \begin{cases} \frac{\mu_p}{2} [I_1 - 3] + S^*(I_5^D) & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \] (8.36)

and

\[ f_{0_m}(F, J, D) = \begin{cases} \frac{\mu_0}{2} [I_1 - 3] + \frac{1}{2\varepsilon_0} I_5^D & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \]

\[ f_{0_p}(F, J, D) = \begin{cases} \frac{\mu_0}{2} [I_1 - 3] + \frac{1}{2\varepsilon_0} I_5^D & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \] (8.37)

Upon substitution of these expressions in the general variational approximation (8.34), it is not difficult to deduce that the optimal comparison medium is such that \( \varepsilon_{0_m} = \varepsilon \), \( \mu_{0_p} = \mu_p \) and, moreover, that
energies (8.37) reads as composite wherein the matrix and filler particles are characterized by the ideal elastic dielectric free
result, it is worth remarking that the macroscopic incompressibility constraint
where the coefficient \( \tilde{\eta} \) from the local incompressibility constraint
\[ \frac{\max_{\mu_0, \varepsilon_0} \{ \tilde{W}_0^*(F, D) + \}}{\min_{\mu_0, \varepsilon_0} \{ \tilde{W}_0^*(F, D) +} \]
\[ (1 - c) \min_{J_1} [\Psi(J_1) - \frac{\mu_0}{2} (J_1 - 3)] + \]
\[ c \min_{J_5} [S^*(J_5) - \frac{1}{2\varepsilon_0} J_5] \}
\[ \frac{\max_{\mu_0, \varepsilon_0} \{ \tilde{W}_0^*(F, D) +} }{\min_{\mu_0, \varepsilon_0} \{ \tilde{W}_0^*(F, D) + } \]
\[ (1 - c) \max_{J_1} [\Psi(J_1) - \frac{\mu_0}{2} (J_1 - 3)] + \]
\[ c \max_{J_5} [S^*(J_5) - \frac{1}{2\varepsilon_0} J_5] \}
\[ + \infty \]
\[ \tilde{W}_0^*(F, D) = \left\{ \begin{array}{ll} \frac{\mu_0}{2} [T_1 - 3] + \frac{1}{2m_{K_0}} \left[ \frac{T_5^D + \tilde{\eta}_0 T_4^D + \tilde{\eta}_0 T_1 T_5^D - T_6^D}{1 + \tilde{\eta}_0^2 + \tilde{\eta}_0 T_2 + \tilde{\eta}_0 T_1} \right] & \text{if } J = 1, \\
+ \infty & \text{otherwise} \end{array} \right. \] (8.39)

where the explicit dependence on \( \tilde{W}_0^* \) on \( F, D \), and \( c \) has been dropped to ease the notation. In this
result, it is worth remarking that the macroscopic incompressibility constraint \( J = 1 \) that ensues
from the local incompressibility constraint \( J = 1 \) in (8.36)–(8.37) is indeed the exact constraint
(within the broad context of comparison medium methods, this is a highly non-trivial result).

Next, we recall from Chapter 7 that the effective free-energy function (8.24) of an elastic dielectric
composite wherein the matrix and filler particles are characterized by the ideal elastic dielectric free
energies (8.37) reads as

\[ \tilde{W}_0^*(F, D) = \left\{ \begin{array}{ll} \frac{\mu_0}{2} [T_1 - 3] + \frac{1}{2m_{K_0}} \left[ \frac{T_5^D + \tilde{\eta}_0 T_4^D + \tilde{\eta}_0 T_1 T_5^D - T_6^D}{1 + \tilde{\eta}_0^2 + \tilde{\eta}_0 T_2 + \tilde{\eta}_0 T_1} \right] & \text{if } J = 1, \\
+ \infty & \text{otherwise} \end{array} \right. \] (8.39)

where the coefficient \( \tilde{\eta}_0 = (\tilde{\varepsilon}_0 - \tilde{m}_{K_0})/\tilde{m}_{K_0} \) has been introduced to ease notation and the effective
coefficients \( \tilde{\mu}_0, \tilde{\varepsilon}_0, \tilde{m}_{K_0} \) are given by

\[ \tilde{\mu}_0 = \frac{1}{5} \int_{\Omega} \left[ (1 - \theta(\mathbf{X})) \mu_0 + \theta(\mathbf{X}) \mu_p \right] K_{klmn} \Gamma_{mkl,n} d\mathbf{X}, \]
\[ \tilde{\varepsilon}_0 = \int_{\Omega} \left[ (1 - \theta(\mathbf{X})) \varepsilon + \theta(\mathbf{X}) \varepsilon_0 \right] \gamma_{m,m} d\mathbf{X}, \]
\[ \tilde{m}_{K_0} = \frac{1}{5} \int_{\Omega} \left[ (1 - \theta(\mathbf{X})) \varepsilon + \theta(\mathbf{X}) \varepsilon_0 \right] K_{ijkl} \Gamma_{rij,s} K_{rstuv} \gamma_{u,k} \gamma_{v,j} d\mathbf{X}. \] (8.40)

In these formulae, explicit use has been made of the optimality conditions \( \varepsilon_0 = \varepsilon \) and \( \mu_0 = \mu_p \),
\( K_{ijkl} = 1/2(\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) - 1/3 \delta_{ij} \delta_{kl} \), and the tensor fields \( \Gamma \) and \( \gamma \) are defined as the solutions of
the following uncoupled linear boundary value problems:

\[
\begin{cases}
\left(\left[\mu_0 + \theta(X)\mu_\gamma\right] \kappa_{ijmn} \Gamma_{mkl,n} + \frac{1}{2} \delta_{ij} q_{kl}\right]_{ij} = 0 & \text{for } X \in \Omega, \quad \Gamma_{ikl} = \delta_{ik} X_l \text{ for } X \in \partial \Omega \\
\Gamma_{mkl,m} = 0
\end{cases}
\]

and

\[
\left(\left[\mu_0 + \theta(X)\varepsilon_{0_p}\right] \gamma_{i,j}\right]_{ij} = 0 \quad \text{for } X \in \Omega, \quad \gamma_i = X_i \text{ for } X \in \partial \Omega.
\]

(8.41)

Granted the result (8.39) for the effective free-energy function of the comparison medium, irrespectively of the growth conditions of the functions \(\Psi\) and \(S^*\), the variational approximation (8.38) can be shown to reduce to

\[
W^* (F, D, c) = \begin{cases}
(1 - c)\Psi(\mathcal{J}_1) - \frac{\mu_0 (1 - c)}{2} [\mathcal{J}_1 - 3] + \frac{\mu_0}{2} [T_1 - 3] + \\
c S^*(\mathcal{J}_5) - \frac{c}{2 \varepsilon_{0_p}} \mathcal{J}_5 + \frac{1}{2m_K} \left[ T_5^D + \eta_0 T_4^D + \eta_0 [T_1 T_5^D - T_6^D] \right] + 3 & \text{if } \mathcal{J} = 1 \\
+ \infty & \text{otherwise}
\end{cases}
\]

(8.43)

with

\[
\mathcal{J}_1 = \frac{1}{1 - c} \left[ (T_1 - 3) \frac{\partial \mu_0}{\partial \mu} - \frac{1}{m_K} \frac{\partial m_K}{\partial \mu} \left( \frac{3T_5^D \eta_0^2 + 2 \eta_0 [T_1 T_5^D + T_4^D - T_6^D] + T_1 T_5^D + T_5^D - T_6^D}{1 + \eta_0^3 + \eta_0^2 T_2 + \eta_0 T_1} \right) \left( \eta_0 T_2 + \eta_0 T_1 \right)^2 + 3 \right]
\]

(8.44)

and

\[
\mathcal{J}_5 = \frac{\varepsilon_{0_p}^2}{c m_K} \frac{\partial m_K}{\partial \varepsilon_{0_p}} \left[ T_5^D + \eta_0^2 \eta_0^3 + \eta_0 [T_1 T_5^D - T_6^D] \right] + \frac{\varepsilon_{0_p}^2}{c m_K} \left( 1 + \eta_0 \right) \frac{\partial m_K}{\partial \varepsilon_{0_p}} - \frac{\partial \varepsilon_{0_p}}{\partial \varepsilon_{0_p}} x \times
\]

\[
\frac{T_1 T_5^D - T_6^D + 2 \eta_0 T_4^D}{1 + \eta_0^3 + \eta_0^2 T_2 + \eta_0 T_1} - \left( T_5^D + \eta_0^2 T_4^D + \eta_0 [T_1 T_5^D - T_6^D] \right) \left( 1 + 2 \eta_0 T_2 + 3 \eta_0^2 \right) \left( 1 + \eta_0^3 + \eta_0^2 T_2 + \eta_0 T_1 \right)^2,
\]

(8.45)

where the variables \(\mu_0\) and \(\varepsilon_{0_p}\) are defined implicitly as the solution of the following system of two nonlinear algebraic equations:

\[
\mathcal{F}_1^* \{ \mu_0, \varepsilon_{0_p} \} \cong \Psi'(\mathcal{J}_1) - \frac{\mu_0}{2} = 0, \quad \mathcal{F}_2^* \{ \mu_0, \varepsilon_{0_p} \} \cong S^*(\mathcal{J}_5) - \frac{1}{2 \varepsilon_{0_p}} = 0.
\]

(8.46)
Having obtained the approximation (8.43) for the effective Helmholtz free energy of the composite, the final step of the derivation consists in computing its Legendre transform (8.35). After making use of the stationarity conditions \( \partial W^\ast / \partial J_1 = \partial W^\ast / \partial J_5 = \partial W^\ast / \partial \mu_0 = \partial W^\ast / \partial \varepsilon_0 = 0 \) and the convexity properties (8.13) of the function \( S^\ast \), the result can be written as

\[
W(\mathbf{F}, \mathbf{E}, c) = \begin{cases} 
(1 - c) \psi(I_1) - \frac{\mu_0 (1 - c)}{2} [I_1 - 3] + \frac{\mu_0}{2} [I_1 - 3] - 
\c S(I_5) + \frac{c \varepsilon_0}{2} I_5 + \frac{\tilde{m}_{K_0} - \varepsilon_0}{2} I_4 - \frac{\tilde{m}_{K_0} - \varepsilon_0}{2} I_5 & \text{if } J = 1 \end{cases}
\]

(8.47)

with

\[
I_1 = \frac{1}{1 - c} \frac{\partial \tilde{\mu}_0}{\partial \tilde{\mu}_0} (I_1 - 3) + \frac{1}{1 - c} \frac{\partial \tilde{m}_{K_0}}{\partial \tilde{\mu}_0} (I_4^E - I_5^E) + 3
\]

and

\[
I_5 = - \frac{1}{c} \left( \frac{\partial \tilde{m}_{K_0}}{\partial \varepsilon_0} - \frac{\partial \varepsilon_0}{\partial \varepsilon_0} \right) I_4 + \frac{1}{c} \frac{\partial \tilde{m}_{K_0}}{\partial \varepsilon_0} I_5^E,
\]

(8.48)

where, again, \( I_1, I_4^E, I_5^E \) stand for the standard (\( \mathbf{F}, \mathbf{E} \))-based invariants defined by (8.15), \( \tilde{\mu}_0, \tilde{\varepsilon}_0, \tilde{m}_{K_0} \) are the effective elastic dielectric coefficients of the comparison medium, as defined by relations (8.40) with (8.41)–(8.42), while the variables \( \mu_0 \) and \( \varepsilon_0 \) are defined implicitly as the solution of the system of two nonlinear algebraic equations

\[
\mathcal{F}_1(\mu_0, \varepsilon_0) \doteq \psi'(I_1) - \frac{\mu_0}{2} = 0, \quad \mathcal{F}_2(\mu_0, \varepsilon_0) \doteq S'(I_5) - \frac{\varepsilon_0}{2} = 0.
\]

(8.49)

Expression (8.47) constitutes the main analytical result of this chapter: it provides a variational approximation for the effective free energy (2.18) of a non-Gaussian dielectric elastomer, with free-energy function (8.1), filled with any type of non-percolative isotropic distribution of nonlinear elastic dielectric particles, with free-energy function (8.5). The following remarks are in order.

i. The macroscopic constitutive response. In view of the equivalence between equations (8.48)–(8.49) and the stationarity conditions \( \partial W / \partial I_1 = \partial W / \partial I_5 = \partial W / \partial \mu_0 = \partial W / \partial \varepsilon_0 = 0 \), the macroscopic electromechanical constitutive relation (2.17) implied by the effective free energy (8.47) is given by

\[
\mathbf{S} = \tilde{\mu}_0 \mathbf{F} + \tilde{m}_{K_0} \mathbf{F}^{-T} \mathbf{E} \otimes \mathbf{F}^{-1} \mathbf{F}^{-T} \mathbf{E} - p \mathbf{F}^{-T},
\]

(8.50)

where \( p \) stands for the arbitrary hydrostatic pressure associated with the incompressibility constraint \( J = 1 \), and

\[
\mathbf{D} = (\varepsilon_0 - \tilde{m}_{K_0}) \mathbf{E} + \tilde{m}_{K_0} \mathbf{F}^{-1} \mathbf{F}^{-T} \mathbf{E}.
\]

(8.51)
Much like the computation of the effective free energy (8.47) itself, the computation of the macroscopic stress (8.50) and macroscopic electric displacement (8.51) for a given microstructure (i.e., a given indicator function $\theta(X)$), given constitutive relations for the matrix and particles (i.e., given $\Psi(I_1), \varepsilon_m, \mu_p,$ and $S(I_5)$), and given macroscopic deformation gradient $\mathbf{F}$ and macroscopic electric field $\mathbf{E}$ amounts to solving the uncoupled linear pdes (8.41)–(8.42) for the fields $\Gamma$ and $\gamma$, evaluating the integrals (8.40) to determine the effective coefficients $\tilde{\mu}_0, \tilde{\varepsilon}_0, \tilde{m}_{K_0}$, and solving the system of nonlinear algebraic equations (8.49) for $\mu_0$ and $\varepsilon_0$.

**ii. The limit of small deformations and moderate electric fields.** By construction, the effective free energy (8.47) is asymptotically exact in the classical limit of small deformations and moderate electric fields. Indeed, in the limit when $\mathbf{F} \to \mathbf{I}$ and $\mathbf{E} \to \mathbf{0}$, the nonlinear algebraic equations (8.49) admit the explicit solution $\mu_0 = 2\Psi'(3) = \mu$ and $\varepsilon_0 = 2S'(0) = \varepsilon_p$ to leading order, and the effective free energy (8.47) reduces asymptotically to (see result (7.7) in Chapter 7)

$$\tilde{W}(\mathbf{F}, \mathbf{E}, c) = \begin{cases} 
2\tilde{\mu} \left[ (\bar{\lambda}_1 - 1)^2 + (\bar{\lambda}_2 - 1)^2 + (\bar{\lambda}_3 - 1)^2 \right] - \\
\frac{\tilde{\varepsilon}}{2} \left[ \mathbf{E}_1^2 + \mathbf{E}_2^2 + \mathbf{E}_3^2 \right] + \\
\tilde{m}_K \left[ (\bar{\lambda}_1 - 1) \mathbf{E}_1^2 + (\bar{\lambda}_2 - 1) \mathbf{E}_2^2 + (\bar{\lambda}_3 - 1) \mathbf{E}_3^2 \right] & \text{if } \bar{\lambda}_1 \bar{\lambda}_2 \bar{\lambda}_3 = 1 \\
\infty & \text{otherwise}
\end{cases}. \quad (8.52)$$

Here, we recall that $\bar{\lambda}_1, \bar{\lambda}_2, \bar{\lambda}_3$ stand for the singular values of the macroscopic deformation gradient tensor $\mathbf{F}, \bar{\mathbf{E}}_1, \bar{\mathbf{E}}_2, \bar{\mathbf{E}}_3$ are the components of the macroscopic electric field $\mathbf{E}$ with respect to any frame of reference of choice, and the effective shear modulus $\tilde{\mu}$, effective permittivity $\tilde{\varepsilon}$, and effective electrostrictive constant $\tilde{m}_K$ are defined by relations (8.40) with (8.41)–(8.42) when evaluated at the aforementioned solution of equations (8.49), namely, $\mu_0 = \mu$ and $\varepsilon_0 = \varepsilon_p$.

**iii. Independence of $\tilde{W}$ on the invariants $T_2$ and $T_6^E$.** For finite deformations and finite electric fields, the effective free energy (8.47) is in general not exact. The direct comparisons with FE simulations presented further below in Section 8.4 support that it does provide, however, an accurate approximation. In this regard, we remark in particular that the effective free energy (8.47) is independent of the invariants $T_2$ and $T_6^E$. The FE simulations presented in Section 8.4 indicate that this distinctive functional trait is indeed exhibited by the corresponding exact solutions.

**iv. The case of ideal elastic dielectric constituents.** For finite deformations and finite electric fields,
the nonlinear algebraic equations (8.49) do not generally admit explicit solutions. There are, nonetheless, a number of special cases of practical interest for which they do. The first one that we report here corresponds to the basic case when both the matrix and the particles are ideal elastic dielectrics, which was the focus of Chapter 7. Namely, when

\[ \Psi(I_1) = \frac{\mu}{2}[I_1 - 3] \quad \text{and} \quad S(I_5^E) = \frac{\varepsilon_p}{2} I_5^E, \]  

(8.53)

the equations (8.49) are solved by \( \mu_0 = \mu \) and \( \varepsilon_{0p} = \varepsilon_p \). In this case, the effective free energy (8.47) reduces, of course, to the result (7.26) obtained in Chapter 7:

\[ W(F,E,c) = \begin{cases} \frac{\bar{\mu}}{2} [T_1^E - 3] + \frac{\bar{m}_K - \bar{\varepsilon}}{2} T_4^E - \frac{\bar{m}_K}{2} T_5^E \quad & \text{if } J = 1 \\ +\infty \quad & \text{otherwise} \end{cases}, \]  

(8.54)

where, as in the asymptotic result (8.52), the effective constants \( \bar{\mu}, \bar{\varepsilon}, \bar{m}_K \) are defined by relations (8.40) with (8.41)–(8.42) when \( \mu_0 = \mu \) and \( \varepsilon_{0p} = \varepsilon_p \).

v. The limiting case of rigid particles: \( \mu_p = +\infty \). The majority of existing experimental investigations on dielectric elastomer composites involve filler particles that are mechanically much stiffer than the underlying elastomeric matrix. For example, PANI, O-CuPc, and TiO\(_2\) particles, which have been often utilized in experimental investigations (see, e.g., Li et al., 2004; Huang et al., 2005; Liu et al., 2013), all exhibit initial shear moduli that are several orders of magnitude larger than the shear moduli of typical dielectric elastomers, such as for instance acrylic elastomers and silicones. This prompts the analysis of the effective free energy (8.47) in the limit of rigid particles when \( \mu_p = +\infty \).

Now, for the case of rigid particles when \( \mu_p = +\infty \), the effective shear modulus \( \bar{\mu}_0 \) of the comparison medium is necessarily linear in \( \mu_0 \), while the effective electrostrictive coefficient \( \bar{m}_{K_0} \) is independent of \( \mu_0 \). More specifically, we have that

\[ \bar{\mu}_0 = (1 - c)r(c)\mu_0 \quad \text{and} \quad \frac{\partial \bar{m}_{K_0}}{\partial \mu_0} = 0 \]  

(8.55)

with

\[ r(c) = \frac{1}{5(1 - c)} \int_\Omega [1 - \theta(X)] K_{klmn} \Gamma_{mkl,n} dX, \]  

(8.56)

where the field \( \Gamma \) here corresponds to the solution of the linear boundary value problem (8.41) for the choice of particle shear modulus \( \mu_p = +\infty \). It follows from (8.55) that equation (8.49)
can be solved in closed form for \( \mu_0 \). The result reads as \( \mu_0 = 2\Psi' \left( r(c) [T_1 - 3] + 3 \right) \). In turn, the effective free energy (8.47) simplifies to

\[
W(F, E, c) = \begin{cases} 
(1 - c)\Psi \left( r(c) [T_1 - 3] + 3 \right) - cS(I_5) + \frac{c \varepsilon_0 I_5}{2} + \frac{\bar{m}K_0}{2} \frac{E^E}{I_4} - \frac{\bar{m}K_0}{2} \frac{T_E}{I_5} & \text{if } J = 1, \\
+\infty & \text{otherwise}
\end{cases},
\]

where we recall that the variable \( I_5 \) is given by expression (8.48), \( \varepsilon_0 \) and \( \bar{m}K_0 \) are the effective permittivity and electrostrictive coefficient of the comparison medium, as defined by relations (8.40) and (8.41)−(8.42) for the case when \( \mu_0 = 2\Psi' \left( r(c) [T_1 - 3] + 3 \right) \) and \( \mu_p = +\infty \), while the variable \( \varepsilon_0 \) is defined implicitly as the solution of the nonlinear algebraic equation (8.49).

In the absence of an electric field when \( E = 0 \), it is fitting to mention that equation (8.49) is solved by \( \varepsilon_0 = \varepsilon_p \) and the effective free-energy function (8.57) reduces to an earlier result of Lopez-Pamies et al. (2013b) for the effective stored-energy function of an isotropic suspension of rigid inclusions in non-Gaussian rubber:

\[
W(F, 0, c) = \begin{cases} 
(1 - c)\Psi \left( r(c) [T_1 - 3] + 3 \right) & \text{if } J = 1, \\
+\infty & \text{otherwise}
\end{cases}. \tag{8.58}
\]

### vi. The limiting case of liquid-like particles: \( \mu_p = 0 \).

Recent theoretical (see, e.g., Lopez-Pamies, 2014; Chapter 6) and experimental (Fassler and Majidi, 2015) investigations have indicated that the addition of liquid-like filler particles — that is, incompressible particles of vanishingly small shear resistance — to dielectric elastomers may have the potential to lead to significantly enhanced elastic dielectric properties.

In the present context, liquid-like particles can be modelled by setting \( \mu_p = 0 \). In this limiting case, much like for rigid particles, the effective shear modulus \( \bar{\mu}_0 \) of the comparison medium can be shown to depend linearly on \( \mu_0 \), while the effective electrostrictive coefficient \( \bar{m}K_0 \) is independent altogether of the value of \( \mu_0 \). In particular, we have that

\[
\bar{\mu}_0 = (1 - c)\varphi(c) \mu_0 \quad \text{and} \quad \frac{\partial \bar{m}K_0}{\partial \mu_0} = 0 \tag{8.59}
\]

with

\[
\varphi(c) = \frac{1}{5(1 - c)} \int_{\Omega} [1 - \theta(X)] K_{klmn} \Gamma_{mkl,n} dX, \tag{8.60}
\]
where the field $\mathbf{\Gamma}$ in this last expression corresponds to the solution of the linear boundary value problem (8.41) for the choice of particle shear modulus $\mu_p = 0$. Given relations (8.59), it is a simple matter to recognize that equation (8.49) can be solved in closed form for the variable $\mu_0$. The result reads as $\mu_0 = 2\Psi' \left( l(c)[I_1 - 3] + 3 \right)$. This in turn leads to the following simplification of the effective free energy (8.47):

$$W(\mathbf{F}, \mathbf{E}, c) = \begin{cases} (1 - c)\Psi \left( l(c) \left[ I_1 - 3 \right] + 3 \right) - c S(I_5) + \frac{c \varepsilon_0 I_5}{2} & \text{if } J = 1, \\ \frac{m_{K_o} - \bar{\varepsilon}_0}{2} T_F - \frac{m_{K_o}}{2} T'_F + \infty & \text{otherwise} \end{cases}$$

where, again, the variable $I_5$ is given by expression (8.48) and $\bar{\varepsilon}_0$ and $m_{K_o}$ are the effective permittivity and electrostrictive coefficient of the comparison medium, as defined by relations (8.40) with (8.41) for the case when $\mu_0 = 2\Psi' \left( l(c)[I_1 - 3] + 3 \right)$ and $\mu_p = 0$, while the variable $\varepsilon_{0_b}$ is defined implicitly by equation (8.49).

vii. The $\mathbf{F}$ and $\mathbf{D}$ formulation. With help of the effective Helmholtz free energy (8.43) and relations (A.4), it is straightforward to partly invert the macroscopic constitutive relations (8.50)–(8.51) in order to rewrite them with the electric displacement field $\mathbf{D}$ playing the role of electric independent variable instead of the electric field $\mathbf{E}$. The result reads as

$$\mathbf{D} = \mu_0 \mathbf{F} - \eta \mathbf{F}^{-T} - \frac{m_{K_o}^3}{m_{K_o}^3 + \eta_0^2 I_2 + \eta_0 I_1} \left[ \mathbf{F} - \eta_0 \mathbf{F}^{-T} \right] + \frac{1}{m_{K_o}^3 \left[ 1 + \eta_0^2 I_2 + \eta_0 I_1 \right]} \times \left[ \eta_0 \left[ I_5^D \mathbf{F} - \mathbf{F} \mathbf{D} \otimes \mathbf{F}^T \mathbf{F} \mathbf{D} - \mathbf{F} \mathbf{F}^T \mathbf{D} \otimes \mathbf{D} \right] + \right]$$

$$\left( 1 + I_1 \eta_0 \right) \mathbf{F} \mathbf{D} \otimes \mathbf{D}$$

where we recall that $\eta_0 = (\bar{\varepsilon}_0 - m_{K_o}^3)/m_{K_o}^3$, $\eta$ stands for the arbitrary hydrostatic pressure associated with the incompressibility constraint $J = 1$, and

$$\mathbf{E} = \frac{m_{K_o}}{m_{K_o}^3 + m_{K_o}^3 I_2 + m_{K_o} I_1} \left[ \left( 1 + I_1 \eta_0 \right) \mathbf{F}^T \mathbf{F} \mathbf{D} + \eta_0^2 \mathbf{D} - \eta_0 \mathbf{F}^T \mathbf{F} \mathbf{F}^T \mathbf{D} \right].$$

viii. Material instabilities. In addition to facilitating the computation of the macroscopic electromechanical constitutive response (8.62)–(8.63), the effective Helmholtz free-energy function (8.43) in terms of $\mathbf{F}$ and $\mathbf{D}$ provides the means to conveniently determine the possible onset of two classes of material instabilities: i) instabilities associated with electromechanical limit loads and ii) microstructural instabilities of long wavelength. Section 7.3.2 discusses the relevant
conditions that signal the triggering of these instabilities and hence we do not repeat them here.

As already indicated above, the variational solution (8.47) applies to arbitrary non-percolative isotropic distributions of filler particles. With the two-pronged objective of demonstrating its use and of facilitating comparisons with experimental results, in the next two subsections we present the specialization of the approximation (8.47) to two specific types of isotropic distributions of filler particles: i) an isotropic iterative microstructure wherein the particles are infinitely polydisperse in size and ii) an isotropic distribution of monodisperse spherical particles. For conciseness, we shall restrict attention to the practically relevant cases of rigid and liquid-like filler particles.

8.2.1 An isotropic iterative microstructure with infinitely polydisperse particles

We begin with the specialization of (8.47) to the iterative microstructure of Lopez-Pamies (2014), wherein the filler particles are infinitely polydisperse in size. In Chapter 3, we worked out exact closed-form solutions for the effective elastic dielectric tensors that characterize the response of dielectric elastomer composites with such iterative microstructures in the limit of small deformations and moderate electric fields. In its general form, this solution applies to anisotropic microstructures, as well as to compressible and anisotropic matrix and filler particle behaviors. In the sequel, we invoke its specialization to the two cases of interest here: incompressible matrix materials and isotropic distributions of rigid and liquid-like particles.

**Rigid particles.** When specialized to incompressible dielectric elastomers with initial shear modulus \( \mu_0 \) and initial permittivity \( \varepsilon \) and to isotropic distributions of rigid particles with initial shear modulus \( \mu_p = +\infty \) and initial permittivity \( \varepsilon_{0p} \), the results (3.56)\(_{1,3,4} \) for the effective elastic dielectric coefficients (8.40) reduces to

\[
\begin{align*}
\tilde{\mu}_0 &= \mu_0 + \frac{5c}{2(1-c)} \mu_0, \\
\tilde{\varepsilon}_0 &= \varepsilon + \frac{3c(\varepsilon_{0p} - \varepsilon)}{(2 + c)\varepsilon + (1 - c)\varepsilon_{0p}} \varepsilon, \\
\tilde{m}_K &= \varepsilon + \frac{3c(\varepsilon_{0p} - \varepsilon)[(23 + 7c)\varepsilon + 7(1 - c)\varepsilon_{0p}]}{10[(2 + c)\varepsilon + (1 - c)\varepsilon_{0p}]^2} \varepsilon.
\end{align*}
\tag{8.64}
\]
It is trivial to deduce from (8.64) that the corresponding function \( r(c) \) defined by (8.56) is thus given by
\[
\begin{align*}
r(c) &= \frac{2 + 3c}{2(1 - c)^2}. 
\end{align*}
\]
Moreover, it is a simple matter to deduce that the finite branch of the effective free-energy function (8.57) for this class of dielectric elastomer composites specializes to
\[
\begin{align*}
W(F, E, c) &= (1 - c)\Psi\left(\frac{2 + 3c}{2(1 - c)^2}[T_1 - 3] + 3\right) - cS(I_5^{H_{1r}}) + \frac{c\varepsilon_0}{2}I_5^{H_{1r}} + \frac{\tilde{m}_K_0 - \varepsilon_0}{2}T_4^E - \frac{\tilde{m}_K_0}{2}T_5^E,
\end{align*}
\]
where \( \varepsilon_0 \) and \( \tilde{m}_K_0 \) are given by expressions (8.64)\_2,\_3,
\[
I_5^{H_{1r}} = \frac{27(1 - c)(\varepsilon_0 - \varepsilon)^2}{\varepsilon_0} + \frac{9[(13 + 2c)\varepsilon + 2(1 - c)\varepsilon_0]\varepsilon^2}{\varepsilon_0^3}T_5^E, 
\]
and the variable \( \varepsilon_0 \) is defined as the solution of the nonlinear algebraic equation
\[
S'(I_5^{H_{1r}}) - \frac{\varepsilon_0}{2} = 0, 
\]
which, in general, for most choices of the function \( S \), needs to be solved numerically.

The effective free-energy function (8.66) contains several limiting cases of practical significance. Here, we report the case corresponding to filler particles that, in addition to being infinitely stiff, are electrically conducting so that the function \( S(I_5^E) = 0 \) if \( I_5^E = 0 \) and \( S(I_5^E) = +\infty \) otherwise.

For such a limiting case, the solution of equation (8.68) can be shown to be given by \( \varepsilon_0 = +\infty \) and the effective free-energy function (8.66) simplifies to the following fully explicit expression:
\[
\begin{align*}
W(F, E, c) &= (1 - c)\Psi\left(\frac{2 + 3c}{2(1 - c)^2}[T_1 - 3] + 3\right) - \frac{9c\varepsilon}{20(1 - c)}T_4^E - \frac{(10 + 11c)\varepsilon}{20(1 - c)}T_5^E. 
\end{align*}
\]

The result (8.69) is relevant for dielectric elastomer composites wherein the filler particles are hard (semi-)conducting polymers (e.g., PANI and O-CuPc) or metals.

**Liquid-like particles.** When specialized to incompressible dielectric elastomers with initial shear modulus \( \mu_0 \) and initial permittivity \( \varepsilon \) and to isotropic distributions of liquid-like particles with initial shear modulus \( \mu_p = 0 \) and initial permittivity \( \varepsilon_0 \), the results (3.56)\_1,\_3,\_4 for the effective elastic dielectric coefficients (8.40) reduces to
\[
\begin{align*}
\tilde{m}_0 &= \mu_0 - \frac{5c}{3 + 2c}\mu_0, \quad \tilde{\varepsilon}_0 = \varepsilon + \frac{3c(\varepsilon_0 - \varepsilon)}{(2 + c)\varepsilon + (1 - c)\varepsilon_0}\varepsilon, \\
\tilde{m}_K_0 &= \varepsilon + \frac{3c(\varepsilon_0 - \varepsilon)[(42 + 26c + 7c^2)\varepsilon + (1 - c)(3 + 7c)\varepsilon_0]}{5(3 + 2c)(2 + c)\varepsilon + (1 - c)\varepsilon_0^2}\varepsilon. 
\end{align*}
\]
The corresponding function \( l(c) \) defined by (8.60) is given by

\[
l(c) = \frac{3}{3 + 2c}.
\]  

In view of the above expressions, the finite branch of the effective free-energy function (8.61) for this class of dielectric elastomer composites reduces to

\[
W(F, E, c) = (1 - c)\Psi \left( \frac{3}{3 + 2c} [I_1 - 3] + 3 \right) - c S \left( I_5^{HJ_1} \right) + \frac{c \varepsilon_0 \sigma_{HJ_1}}{2} + \frac{\overline{m}_{K_0} - \overline{\varepsilon}_0}{2} T_4^E - \frac{\overline{m}_{K_0}}{2} T_5^E,
\]

where \( \overline{\varepsilon}_0 \) and \( \overline{m}_{K_0} \) are given by expressions (8.70),

\[
I_5^{HJ_1} = \frac{54(1 - c)(4 + c)(\varepsilon_0 - c)\varepsilon^2}{5(3 + 2c)[(2 + c)\varepsilon + (1 - c)\varepsilon_0]^3} T_4^E + \frac{9((54 + 17c + 4c^2)\varepsilon - (1 - c)(9 - 4c)\varepsilon_0)\varepsilon^2}{5(3 + 2c)[(2 + c)\varepsilon + (1 - c)\varepsilon_0]^3} T_5^E,
\]

and \( \varepsilon_0 \) is defined implicitly by the nonlinear algebraic equation

\[
S' \left( I_5^{HJ_1} \right) - \frac{\varepsilon_0}{2} = 0,
\]

which, similar to (8.68), for most choices of the function \( S \), needs to be solved numerically.

For the limiting case when the particles, in addition to being of vanishingly small shear resistance, are electrically conducting, the solution of equation (8.74) can be shown to be given by \( \varepsilon_0 = +\infty \) and the effective free-energy function (8.72) reduces to

\[
W(F, E, c) = (1 - c)\Psi \left( \frac{3}{3 + 2c} [I_1 - 3] + 3 \right) - \frac{9c(4 + c)\varepsilon}{10(3 - c - 2c^2)} T_4^E - \frac{(15 + 4c + 11c^2)\varepsilon}{10(3 - c - 2c^2)} T_5^E.
\]

The simple and fully explicit result (8.75) is relevant for dielectric elastomer composites wherein the filler particles are conducting liquids (e.g., Galinstan).

### 8.2.2 An isotropic distribution of monodisperse spherical particles

Complementary to the foregoing results, in this subsection we present the specialization of the effective free energy (8.47) to an isotropic distribution of monodisperse spherical particles.

Now, for an isotropic distribution of monodisperse spherical particles, the pdes (8.41) and (8.42) do not admit an analytical solution. As discussed in Chapter 5, however, they can be readily solved numerically by means of finite elements (FE). This in turn allows for the numerical evaluation of the effective coefficients (8.40) required in the computation of the effective free energy (8.47). In general, since the derivatives \( \partial \overline{\mu}_0 / \partial \mu_0, \partial \overline{m}_{K_0} / \partial \mu_0, \partial \overline{\varepsilon}_0 / \partial \varepsilon_0, \partial \overline{m}_{K_0} / \partial \varepsilon_0 \) of the effective coefficients (8.40) are also needed in the computation of (8.47), the pdes (8.41) and (8.42) need to be solved multiple
times for a sufficiently wide range of values $\mu_0 > 0$ and $\varepsilon_{0_p} \geq \varepsilon_0$ to make possible the numerical computation of these derivatives. For the two specific cases of interest here when the filler particles are either rigid or liquid-like, the pde (8.41) needs to be solved only once since it is independent of $\mu_0$ (see remarks $v$ and $vi$ above) and only the pde (8.42) needs to be solved multiple times for a sufficiently wide range of values $\varepsilon_{0_p} \geq \varepsilon_0$. For these two cases, moreover, it is possible to obtain from the FE solutions simple explicit interpolating formulas for the effective elastic dielectric coefficients (8.40) over a large range of volume fractions of particles $c$. In the sequel, we report such formulas for the practically relevant range $c \in [0, 0.25]$.

**Rigid particles.** For incompressible dielectric elastomers, with initial shear modulus $\mu_0$ and initial permittivity $\varepsilon$, and rigid spherical particles, with initial shear modulus $\mu_p = +\infty$ and initial permittivity $\varepsilon_{0_p}$, the FE solutions for the effective elastic dielectric coefficients (8.40) are accurately described for the range of volume fraction of particles $c \in (0, 0.25]$ by the following explicit formulas:

\[
\bar{\mu}_0 = \frac{\mu_0}{(1 - c)^{5/2}}, \quad \bar{\varepsilon}_0 = \varepsilon + \frac{3c(\varepsilon_{0_p} - \varepsilon)}{(2 + c)\varepsilon + (1 - c)\varepsilon_{0_p}},
\]

\[
\bar{m}_{K_0} = \varepsilon + \frac{3c(\varepsilon_{0_p} - \varepsilon)[(10 + 2c + 3c^2)\varepsilon + (1 - c)(5 + 3c)\varepsilon_{0_p}]}{5[(2 + c)\varepsilon + (1 - c)\varepsilon_{0_p}]^2}.
\] (8.76)

The corresponding function $r(c)$ defined by (8.56) is given by

\[
r(c) = \frac{1}{(1 - c)^{5/2}}. \quad \text{(8.77)}
\]

In view of relations (8.76)–(8.77), the finite branch of the effective free-energy function (8.57) for this class of dielectric elastomer composites specializes to

\[
\mathcal{W} (F, E, c) = (1-c)\mathcal{W}\left(\frac{T_1 - 3}{(1-c)^{7/2}} + 3\right) - c S \left(\frac{T_5^{\text{Sph}}}{\bar{\varepsilon}_0} + \frac{\bar{m}_{K_0} - \bar{\varepsilon}_0}{2} T_4 - \frac{\bar{m}_{K_0}}{2} T_5^E, \right) \quad (8.78)
\]

where $\bar{\varepsilon}_0$ and $\bar{m}_{K_0}$ are given by expressions (8.76)$_{2,3}$,

\[
T_5^{\text{Sph}} = -\frac{54c(1-c)(\varepsilon_{0_p} - \varepsilon)^2}{5[(2 + c)\varepsilon + (1 - c)\varepsilon_{0_p}]^3} T_4^E + \frac{9[(10 - c + 6c^2)\varepsilon + (5 + c - 6c^2)\varepsilon_{0_p}]\varepsilon^2}{5[(2 + c)\varepsilon + (1 - c)\varepsilon_{0_p}]^3} T_5^E, \quad (8.79)
\]

and the variable $\varepsilon_{0_p}$ is defined implicitly by the nonlinear algebraic equation

\[
S' \left(\frac{T_5^{\text{Sph}}}{\bar{\varepsilon}_0}\right) - \frac{\varepsilon_{0_p}}{2} = 0. \quad (8.80)
\]

For the limiting case when the particles are electrically conducting, equation (8.80) admits the explicit solution $\varepsilon_{0_p} = +\infty$ and the effective free-energy function (8.78) reduces to

\[
\mathcal{W} (F, E, c) = (1-c)\mathcal{W}\left(\frac{T_1 - 3}{(1-c)^{7/2}} + 3\right) + \frac{9c^2\varepsilon}{10(1-c)} T_4^E - \frac{(5 + 10c + 9c^2)\varepsilon}{10(1-c)} T_5^E. \quad (8.81)
\]
Again, much like the effective free-energy function (8.69), the fully explicit effective free-energy function (8.81) is relevant for dielectric elastomer composites wherein the filler particles are hard (semi-)conducting materials.

**Liquid-like particles.** For incompressible dielectric elastomers, with initial shear modulus $\mu_0$ and initial permittivity $\varepsilon$, and liquid-like spherical particles, with initial shear modulus $\mu_p = 0$ and initial permittivity $\varepsilon_0$, the FE solutions for the effective elastic dielectric coefficients (8.40) are accurately described for the range of volume fraction of particles $c \in [0, 0.25]$ by the following formulas:

$$
\tilde{\mu}_0 = (1 - c)^{5/3} \mu_0, \quad \tilde{\varepsilon}_0 = \varepsilon + \frac{3c \varepsilon (\varepsilon_0 - \varepsilon)}{(2 + c) \varepsilon + (1 - c) \varepsilon_0},
$$

$$
\tilde{m}_{K_0} = \varepsilon + \frac{9c(\varepsilon_0 - \varepsilon)^2}{[2 + c] \varepsilon + (1 - c) \varepsilon_0]^{2/3}} \left( \frac{4c^2 - 81c^{61/25}}{500} \varepsilon + \frac{c \varepsilon}{\varepsilon_0 - \varepsilon} \right). \tag{8.82}
$$

The corresponding function $l(c)$ defined by (8.60) is given by

$$
l(c) = (1 - c)^{2/3}. \tag{8.83}
$$

It follows from expressions (8.82)–(8.83) that the finite branch of the effective free-energy function (8.61) for this class of dielectric elastomer composites reduces to

$$
\overline{W}(\overline{F}, \overline{E}, c) = (1 - c) \Psi \left( (1 - c)^{2/3} [I_1 - 3] + 3 \right) - c S \left( \mathcal{I}_5^{\text{Sph}} \right) + \frac{c \varepsilon_0}{2} \mathcal{I}_5^{\text{Sph}} + \frac{\tilde{m}_{K_0} - \tilde{\varepsilon}_0}{2} T_4^E - \frac{\tilde{m}_{K_0}}{2} T_5^E, \tag{8.84}
$$

where $\tilde{\varepsilon}_0$ and $\tilde{m}_{K_0}$ are given by expressions (8.82)_{2,3},

$$
\mathcal{I}_5^{\text{Sph}} = \frac{3(1500 - 1900c + 729c^{36/25}) (\varepsilon_0 - \varepsilon)^2}{250([2 + c] \varepsilon + (1 - c) \varepsilon_0)^3} T_4^E - \frac{3([750 - 1150c + 729c^{36/25}] (\varepsilon_0 - \varepsilon) - 2250 \varepsilon)^2}{250([2 + c] \varepsilon + (1 - c) \varepsilon_0)^3} T_5^E, \tag{8.85}
$$

and $\varepsilon_0$ is implicitly defined by the nonlinear algebraic equation

$$
S' \left( \mathcal{I}_5^{\text{Sph}} \right) - \frac{\varepsilon_0}{2} = 0. \tag{8.86}
$$

For the limiting case when the particles, in addition to being of vanishingly small shear resistance, are electrically conducting, the solution of equation (8.86) is simply given by $\varepsilon_0 = +\infty$ and the effective free-energy function (8.84) simplifies to

$$
\overline{W}(\overline{F}, \overline{E}, c) = (1 - c) \Psi \left( (1 - c)^{2/3} [I_1 - 3] + 3 \right) - \frac{c(1500 - 1900c + 729c^{36/25}) \varepsilon}{1000(1 - c)^2} T_4^E - \frac{(500 - 1000c + 900c^2 - 729c^{61/25}) \varepsilon}{1000(1 - c)^2} T_5^E. \tag{8.87}
$$
8. Nonlinear electroelastic deformations of DECs: Non-Gaussian elastic dielectrics

Akin to the effective free-energy function (8.75), the fully explicit effective free-energy function (8.87) is relevant for dielectric elastomer composites wherein the fillers are conducting liquids.

8.2.3 Electrostriction

We conclude this section by spelling out the response predicted by the effective free-energy function (8.47) for the experimentally prominent boundary conditions of uniaxial electrostriction when the macroscopic first Piola-Kirchhoff stress $\mathbf{S}$ and macroscopic electric field $\mathbf{E}$ are of the form

$$
\mathbf{S}_{ij} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \mathbf{E}_i = \begin{bmatrix} 0 \\ 0 \\ E \end{bmatrix}; \quad (8.88)
$$

the components of all tensorial quantities throughout this subsection are referred to the Cartesian laboratory axes $e_1, e_2, e_3$ depicted in Fig. 8.1. It follows from the constitutive relations (8.50) and (8.51) that

$$
F_{ij} = \begin{bmatrix} \lambda^{-1/2} & 0 & 0 \\ 0 & \lambda^{-1/2} & 0 \\ 0 & 0 & \lambda \end{bmatrix} \quad \text{and} \quad D_i = \begin{bmatrix} 0 \\ 0 \\ D \end{bmatrix}, \quad (8.89)
$$

where the electrostriction stretch $\lambda$ in the direction of the applied electric field (see Fig. 8.1) and the non-trivial component $D$ of the electric displacement field are defined by the relations

$$
\lambda^4 - \lambda + \frac{\bar{m}_K \varepsilon_0}{\mu_0} \mathbf{E}^2 = 0 \quad \text{and} \quad D = \left[ \bar{\varepsilon}_0 - \bar{m}_K \left( 1 - \frac{1}{\lambda^2} \right) \right] \mathbf{E} \quad (8.90)
$$

in terms of the applied electric field $\mathbf{E}$. Here, we stress yet again that the coefficients $\bar{\mu}_0, \bar{\varepsilon}_0, \bar{m}_K$ in (8.90) are ultimately functions of the microstructure through the indicator function $\theta(\mathbf{X})$, of the properties of the dielectric elastomeric matrix through the function $\Psi$ and initial permittivity $\varepsilon$, of the properties of the filler particles through the initial shear modulus $\mu_p$ and function $\mathcal{S}$, and of the macroscopic deformation gradient $\mathbf{F}$ and electric field $\mathbf{E}$, in this case, through the electrostriction stretch $\lambda$ and uniaxial component $E$. 
8.2.3.1 The case of an isotropic distribution of monodisperse spherical particles

For demonstration purposes and later use in Section 8.4, we spell out next the specialization of the general results (8.90) to the case discussed in Section 8.2.2 when the microstructure consists of an isotropic distribution of monodisperse spherical particles that are either rigid or liquid-like.

**Rigid particles.** For the case of rigid spherical particles, the coefficients $\tilde{\mu}_0$, $\tilde{\varepsilon}_0$, $\tilde{m}_K_0$ are given by expressions (8.76) so that relations (8.90) defining the electrostriction stretch $\lambda$ and electric displacement $D$ specialize to

$$\lambda^4 - \lambda^3 - \frac{3c(1-c)^{5/2}(\varepsilon_0 - \varepsilon)\varepsilon}{3c(\varepsilon_0 - \varepsilon) + (10 + 2c + 3c^2)\varepsilon + (1-c)(5 + 3c)\varepsilon_0} = 0$$

(8.91)

and

$$D = \varepsilon \left[ \frac{1}{\lambda^2} + \frac{3c(\varepsilon_0 - \varepsilon)}{(2 + c)\varepsilon + (1-c)\varepsilon_0} \left( 1 + \frac{(1-\lambda^2)[(10 + 2c + 3c^2)\varepsilon + (1-c)(5 + 3c)\varepsilon_0]}{5[(2 + c)\varepsilon + (1-c)\varepsilon_0]\lambda^2} \right) \right] E,$$

(8.92)

where $\varepsilon_0$ is solution of the nonlinear algebraic equation

$$\mathcal{S} \left( I_0^{\mathrm{Sph.}} - \frac{\varepsilon_0}{2} \right) = 0$$

(8.93)

with

$$I_0^{\mathrm{Sph.}} = \frac{54c(1-c)(\varepsilon_0 - \varepsilon)^2}{5[(2 + c)\varepsilon + (1-c)\varepsilon_0]^3} E^2 + \frac{9[(10 - c + 6c^2)\varepsilon + (5 + c - 6c^2)\varepsilon_0]^2}{5[(2 + c)\varepsilon + (1-c)\varepsilon_0]^3} \frac{E^2}{\lambda^2}.$$
If, in addition to being rigid, the particles are electrically conducting, the results (8.91)–(8.92) reduce to

\[
\lambda^4 - \lambda + \frac{(1-c)^{3/2}(5+10c+9c^2)\varepsilon}{10\Psi'} \left(\frac{\lambda^2 + 2\lambda^{-1} - 3}{(1-c)^{7/2}} + 3\right) E^2 = 0
\]

(8.95)

and

\[
\mathcal{D} = \frac{[5 + 10c + 9c^2(1-\lambda^2)]\varepsilon}{5(1-c)\lambda^2} E.
\]

(8.96)

**Liquid-like particles.** For the case of liquid-like spherical particles, the coefficients \(\tilde{\mu}_0, \tilde{\varepsilon}_0, \tilde{m}_K_0\) are given by expressions (8.82) so that relations (8.90) defining the electrostriction stretch \(\lambda\) and electric displacement \(\mathcal{D}\) take the form

\[
\lambda^4 - \lambda + \frac{9c\varepsilon(\varepsilon_0 - \varepsilon)^2}{((2+c)\varepsilon + (1-c)\varepsilon_0)^2} \left(\frac{4c^2}{45} - \frac{81c^{61/25}}{500} + \frac{c\varepsilon}{\varepsilon_0 - \varepsilon}\right) E^2 = 0
\]

(8.97)

and

\[
\mathcal{D} = \varepsilon \left[\frac{1}{\lambda^2} + \frac{3c(\varepsilon_0 - \varepsilon)}{(2+c)\varepsilon + (1-c)\varepsilon_0} (1 + \frac{(1-\lambda^4)(\varepsilon_0 - \varepsilon)}{1500[(2+c)\varepsilon + (1-c)\varepsilon_0]\lambda^2})\right] E,
\]

(8.98)

where \(\varepsilon_0\) is implicitly defined by the nonlinear algebraic equation

\[
S' \left(\mathcal{I}_5^{\text{Sph}}\right) - \frac{\varepsilon_0}{2} = 0
\]

(8.99)

with

\[
\mathcal{I}_5^{\text{Sph}} = \frac{3(1500 - 1900c + 729c^{36/25})(\varepsilon_0 - \varepsilon)^2}{250[(2+c)\varepsilon + (1-c)\varepsilon_0]^3} E^2 - \frac{3[(750 - 1150c + 729c^{36/25})(\varepsilon_0 - \varepsilon) - 2250\varepsilon]^2 E^2}{250[(2+c)\varepsilon + (1-c)\varepsilon_0]^3}\lambda^2.
\]

(8.100)

If, in addition to exhibiting a vanishingly small shear resistance, the particles are electrically conducting, the results (8.97)–(8.98) specialize further to

\[
\lambda^4 - \lambda + \frac{(500 - 1000c + 900c^2 - 729c^{61/25})\varepsilon}{1000(1-c)^{11/3}\Psi'} \left((1-c)^{2/3}\lambda^2 + 2\lambda^{-1} - 3\right) + 3) E^2 = 0
\]

(8.101)

and

\[
\mathcal{D} = \frac{[500 - 729c^{61/25}(1-\lambda^2) - 500c(2-3\lambda^2) + 100c^2(9-19\lambda^2)]\varepsilon}{500(1-c)^2\lambda^2} E.
\]

(8.102)
8. A hybrid FE formulation for homogenization problems in nonlinear electroelastostatics

Complementary to the analytical framework put forth in the two preceding sections, we present in this section a hybrid FE formulation to construct homogenization solutions numerically for the macroscopic elastic dielectric response of dielectric elastomer composites subjected to finite deformations and finite electric fields. Non-hybrid FE formulations for this class of problems appear to have been first reported by Li and Landis (2012) and by Keip et al. (2014) in the context of two spatial dimensions, while non-hybrid FE formulations in three dimensions have been just recently reported by Miehe et al. (2016). These works also include sample solutions for square/cubic distributions of elliptical/ellipsoidal distributions of filler particles in compressible ideal elastic dielectric matrix materials under conditions of uniaxial electrostriction. Because of the well-known issue of volumetric locking, the above referenced non-hybrid formulations are not applicable to study incompressible or nearly incompressible dielectric elastomer composites, which are the case of most practical relevance and of interest here.

For definiteness, we restrict attention to dielectric elastomer composites of infinite extent whose initial microstructures are defined by the periodic repetition of a cubic unit cell, $Y = \{ X : X \in [0,1]^3 \}$ say, containing a finite, possibly large, number of filler particles. The hybrid FE formulation that we pursue here stems from the definitions of a function $\hat{W}(X, F, J, E)$ that agrees identically with the local free-energy function $W(X, F, E)$ when $J = \text{det} F$ and of its transform (see, e.g., Chi et al., 2014)

$$\hat{W}^*(X, F, J, E) = \max_J \left\{ p(J - 1) - \hat{W}(X, F, J, E) \right\}.$$  

(8.103)

Provided that $\hat{W}$ is convex in its argument $J$, the duality relation

$$\hat{W}(X, F, J, E) = \max_p \left\{ p(J - 1) - \hat{W}^*(X, F, p, E) \right\}$$  

(8.104)

follows from (8.103). Direct use of (8.104) and restricting attention to $Y$-periodic solutions indicates that one to rewrite the variational problem (2.18) in the alternative form

$$\bar{W}(F, E, c) = \min_{u \in U} \max_{\Phi \in F} \max_{p \in P} \int_Y \left\{ p[\text{det} F(u) - 1] - \hat{W}^*(X, F(u), p, E(\Phi)) \right\} \, dX,$$  

(8.105)

2Periodic solutions of larger period than the unit cell $Y$ may exit. These are associated with the development of microscopic instabilities (see, e.g., Geymonat et al., 1993; Michel et al., 2010). We shall not consider such solutions here.
where the displacement vector field \( \mathbf{u} = \mathbf{x} - \mathbf{X} \) and the electric potential scalar field \( \Phi \), both free of any differential constraint, are conveniently chosen as the independent arguments in lieu of the deformation gradient \( \mathbf{F} \) and the curl-free Lagrangian electric field \( \mathbf{E} \). For clarity, we write the dependence of \( F_{ij}(u_i) = \delta_{ij} + u_{i,j} \) and \( E_i(\Phi) = -\Phi_i \) on \( \mathbf{u} \) and \( \Phi \) explicitly. In the hybrid variational principle (8.105), \( \mathcal{U}, \mathcal{F}, \) and \( \mathcal{P} \) stand for sufficiently large sets of admissible displacements \( \mathbf{u}, \) admissible electric potentials \( \Phi, \) and admissible pressure fields \( p \) that are consistent with the following periodicity conditions:

\[
\mathbf{u}(\mathbf{X}) = (\mathbf{F} - \mathbf{I})\mathbf{X} + \tilde{\mathbf{u}}(\mathbf{X}), \text{ where } \tilde{\mathbf{u}} \text{ is } Y-\text{periodic, } \tilde{\mathbf{u}}(0) = \mathbf{0}, \\
\Phi(\mathbf{X}) = -\mathbf{E} \cdot \mathbf{X} + \tilde{\Phi}(\mathbf{X}), \text{ where } \tilde{\Phi} \text{ is } Y-\text{periodic, } \tilde{\Phi}(0) = 0, \\
p \text{ is } Y-\text{periodic.} (8.106)
\]

The weak form of the Euler-Lagrange equations associated with the variational principle (8.105) reads as

\[
\int_Y \left\{ \left( p \left( \det \mathbf{F}(\mathbf{u}) \right) F^{-1}(\mathbf{u}) - \frac{\partial \hat{W}^*}{\partial F_{ij}}(\mathbf{X}, \mathbf{F}(\mathbf{u}), p, \mathbf{E}(\Phi)) \right) \right\} v_{i,j} \, d\mathbf{X} = 0 \quad \forall v \in \mathcal{U}^0, \\
\int_Y \left\{ \frac{\partial \hat{W}^*}{\partial E_i}(\mathbf{X}, \mathbf{F}(\mathbf{u}), p, \mathbf{E}(\Phi)) \right\} \psi_i \, d\mathbf{X} = 0 \quad \forall \psi \in \mathcal{F}^0, \\
\int_Y \left\{ \det \mathbf{F}(\mathbf{u}) - 1 - \frac{\partial \hat{W}^*}{\partial p}(\mathbf{X}, \mathbf{F}(\mathbf{u}), p, \mathbf{E}(\Phi)) \right\} q \, d\mathbf{X} = 0 \quad \forall q \in \mathcal{P}, (8.107)
\]

where \( \mathcal{U}^0 \) and \( \mathcal{F}^0 \) stand for sufficiently large spaces of vector fields \( \mathbf{v} \) and scalar fields \( \psi \) that are \( Y \)-periodic and satisfy \( \mathbf{v}(0) = \mathbf{0} \) and \( \psi(0) = 0 \).

A convenient yet robust method, which we pursue here, to construct numerical solutions for the type of Euler-Lagrange equations (8.107) is the conforming FE method. This requires the selection of suitable finite dimensional subspaces of \( \mathcal{U}, \mathcal{F}, \) and \( \mathcal{P} \). The details of the construction of such subspaces and of the FE solutions for the Euler–Lagrange equation (8.107) that they allow to generate are provided in Appendix D.4.1.

### 8.3.1 Application to non-Gaussian dielectric elastomers isotropically filled with nonlinear elastic dielectric particles

The hybrid FE formulation presented Appendix D.4.1 is applicable to arbitrary types — in terms of local elastic dielectric behaviors as well as microstructures — of dielectric elastomer composites.
The interest of this chapter is on non-Gaussian dielectric elastomers, characterized by free-energy functions of the form (8.1), isotropically filled with nonlinear elastic dielectric particles characterized by free-energy functions of the form (8.5). For this class of dielectric elastomer composites, the transform function \( \hat{W}^*(X,F,p,E) \) required in the hybrid variational principle (8.105) is simply given by

\[
\hat{W}^*(X,F,p,E) = [1 - \theta(X)]\hat{W}^*_n(F,p,E) + \theta(X)\hat{W}^*_p(F,p,E)
\]  

(8.108)

with

\[
\hat{W}^*_n(F,p,E) = -\Psi(I_1) + \frac{\varepsilon}{2}I_5^E \quad \text{and} \quad \hat{W}^*_p(F,p,E) = -\frac{\mu_p}{2}(I_1 - 3) + S(I_5^E),
\]  

(8.109)

this for any isotropic indicator function \( \theta \) of choice.

Figure 8.2: Meshes for unit cells \( Y \) containing: (a) \( N \approx 36 \) randomly distributed spherical particles of three different sizes with \( N_n \approx 150,000 \) elements (\( N_n \approx 670,000 \) nodes) and (b) \( N \approx 30 \) randomly distributed spherical particles of identical size with \( N_n \approx 80,000 \) elements (\( N_n \approx 345,000 \) nodes). The volume fraction of particles in both meshes is \( c = 0.15 \).

Now, by definition, an isotropic distribution of particles involves an infinite number of particles. Within the present context, however, it suffices to consider a large but finite number of particles randomly distributed in the repeating unit cell \( Y \). In this chapter, we follow this approach to work out FE results for two types of microstructures: i) an isotropic distribution of polydisperse spherical particles of a finite number of different sizes and ii) an isotropic distribution of monodisperse spherical particles, as described in Sections 7.2.1 and 7.2.2. Representative meshes for two realizations of such microstructures are shown in Fig. 8.2 for a composite where the volume fraction of particles is \( c = 0.15 \). Mesh sensitivity analyses have indicated that meshes comprising \( N_n \approx 150,000 \) elements (\( N_n \approx 670,000 \) nodes) for the polydisperse microstructure and \( N_n \approx 80,000 \) elements (\( N_n \approx 345,000 \) nodes) for the monodisperse microstructure are refined enough to deliver accurate results, at least
8. Nonlinear electroelastic deformations of DECs: Non-Gaussian elastic dielectrics

for the range of volume fraction of particles considered in this chapter, \( c \in [0, 0.25] \). Finally, we note that all the FE results presented throughout this chapter correspond to the average of three realizations (see Sections 7.2.1 and 7.2.2). The responses of all three realizations exhibited very small differences (less than 1%) between one another.

### 8.4 Sample results

Next, we present sample results for the macroscopic electromechanical behaviors of a typical non-Gaussian dielectric elastomer filled with several physically significant types of nonlinear elastic dielectric filler particles, as characterized by the analytical variational solution (8.47) and by the hybrid FE formulation of Section 8.3. Throughout this section, for definiteness, we make use of the choice (8.3) for the function \( \Psi(I_1) \) described at the beginning of this chapter, so that

\[
W^{(1)}(F, E) = \left\{ \begin{array}{ll}
\frac{3^{1-\alpha_1}}{2\alpha_1} \mu_1[I_1^{\alpha_1} - 3^{\alpha_1}] + \frac{3^{1-\alpha_2}}{2\alpha_2} \mu_2[I_1^{\alpha_2} - 3^{\alpha_2}] - \frac{\varepsilon_0}{2} I_5^E & \text{if } \det F = 1 \\
+\infty & \text{otherwise}
\end{array} \right.,
\]

with the material parameters listed in Table 8.1. This choice of free-energy function provides an accurate model for a standard silicone elastomer over large ranges of deformations and electric fields; see Section 2.3 in Lopez-Pamies (2010) and Liu et al. (2013).

<table>
<thead>
<tr>
<th>Material</th>
<th>( \alpha_1 )</th>
<th>( \mu_1 ) (MPa)</th>
<th>( \alpha_2 )</th>
<th>( \mu_2 ) (MPa)</th>
<th>( \varepsilon ) (F/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicone elastomer</td>
<td>3.837</td>
<td>0.032</td>
<td>0.559</td>
<td>0.3</td>
<td>3.2 ( \varepsilon_0 )</td>
</tr>
</tbody>
</table>

Table 8.1: Values for the materials parameters \( \alpha_1, \mu_1, \alpha_2, \mu_2, \varepsilon \) in the free-energy function (8.110) fitted to model a silicone elastomer.

Throughout this section, we also make use of the choice (8.10) for the function \( S(I_5^E) \), so that the free-energy function characterizing the response of the particles is given by

\[
W^{(2)}(F, E) = \left\{ \begin{array}{ll}
\frac{\mu_p}{2} [I_1 - 3] - \frac{\varepsilon_0}{2} I_5^E - \frac{p_s^2}{3(\varepsilon_p - \varepsilon_0)} \times \\
\ln \left( \sinh \left( \frac{3(\varepsilon_p - \varepsilon_0) \sqrt{I_5^E}}{p_s} \right) \right) - \ln \left( \frac{3(\varepsilon_p - \varepsilon_0) \sqrt{I_5^E}}{p_s} \right) & \text{if } \det F = 1 \\
+\infty & \text{otherwise}
\end{array} \right.,
\]

(8.111)

In particular, within the context of this model, we consider the sets of initial shear modulus \( \mu_p \), initial permittivity \( \varepsilon_p \), and saturation polarization \( p_s \) that are listed in Table 8.2. These correspond
to the practically relevant cases of rigid conducting particles, rigid high-permittivity particles exhibiting polarization saturation, and liquid-like high-permittivity particles. They are representative of filler particles made out of metals or hard conducting polymers, polarization-saturating ferroelectric ceramics, and high-permittivity liquids, the first two of which have been widely utilized in experiments (see, e.g., Huang and Zhang (2004); Huang et al. (2005); Mc Carthy et al. (2009); Liu et al. (2013)).

<table>
<thead>
<tr>
<th></th>
<th>$\mu_p$ (MPa)</th>
<th>$\varepsilon_p$ (F/m)</th>
<th>$p_s$ (C/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rigid conducting particles</td>
<td>$+\infty$</td>
<td>$+\infty$</td>
<td>$+\infty$</td>
</tr>
<tr>
<td>Rigid high-permittivity particles with polarization saturation</td>
<td>$+\infty$</td>
<td>$320\varepsilon_0$</td>
<td>$10^{-2}$, $10^{-4}$</td>
</tr>
<tr>
<td>Liquid-like high-permittivity particles</td>
<td>0.03</td>
<td>$320\varepsilon_0$</td>
<td>$+\infty$</td>
</tr>
</tbody>
</table>

Table 8.2: Values for the materials parameters $\mu_p$, $\varepsilon_p$, $p_s$ in the free-energy function (8.111) utilized to model rigid conducting particles, rigid high-permittivity particles with polarization saturation, and liquid-like high-permittivity particles.

We begin in Section 8.4.1 by reporting sample results that assess the accuracy of the variational solution (8.47) via direct comparisons with FE results. For conciseness, we restrict attention to microstructures wherein the fillers are monodisperse spherical particles. The analytical solution (8.47) is subsequently deployed to probe the electrostriction response of the above-introduced non-Gaussian silicone elastomer filled with rigid conducting particles (Section 8.4.2), rigid high-permittivity particles with polarization saturation (Section 8.4.3), and liquid-like high-permittivity particles (Section 8.4.4). These latter sets of results are aimed at scrutinizing the effects of the elastic dielectric properties of the underlying fillers, as well as their content and size dispersion on the macroscopic electrostriction properties of promising classes of dielectric elastomer composites.

8.4.1 Accuracy of the variational solution (8.47) at finite deformations and finite electric fields

Figure 8.3 presents comparisons between the variational solution (8.47) and corresponding FE solutions. As for all the results presented in this section, the underlying dielectric elastomeric matrix corresponds to the non-Gaussian silicone elastomer characterized by the free-energy function (8.110) with material parameters given in Table 8.1. The fillers are monodisperse spherical particles whose
elastico-elastic behaviors are characterized by the free-energy function \((8.111)\) with material parameters given in Table 8.2. Specifically, Figs. 8.3(a) and (b) present comparisons for the case of rigid conducting particles with volume fraction \(c = 0.05\). Figures 8.3(c) and (d) show comparisons for rigid high-permittivity particles with polarization saturation \(p_s = 10^{-4}\) C/m\(^2\) at volume fraction \(c = 0.05\). Figures 8.3(e) and (f) pertain to liquid-like high-permittivity particles with volume fraction \(c = 0.15\).

The plots in Fig. 8.3 show the effective free energy \(\tilde{W}\) as a function of the invariants \(I_2\) and \(I_6^E\) for fixed values of the remaining four isotropic invariants, \(I_1, I_4, I_5, I_6\) and \(I_1, I_2, I_4, I_5\). We note that keeping the values of four out of the five invariants \(I_1, I_2, I_4, I_5, I_6\) constant restricts the range of physical values that the remaining free invariant can take on. For instance, for the prescribed set of invariants \(I_1 = 3.86, I_4^E = 7.1 \times 10^3\) MV/m\(^2\), \(I_5^E = 11.7 \times 10^3\) MV/m\(^2\), \(I_6^E = 30.0 \times 10^3\) MV/m\(^2\) in Fig. 8.3(a), the range of physically allowable values for \(I_2\) is \([4.13, 4.24]\).

The results shown in Fig. 8.3 span the entire range of physically allowable values for each one of the cases that is presented. We further note that the selected values of the macroscopic invariants \(I_1, I_2, I_4^E, I_5^E, I_6^E\) in Fig. 8.3 involve local invariants \(I_1(X)\) and \(I_5^E(X)\) that are large enough in the silicone elastomer and in the filler particles so as to trigger the non-Gaussian stiffening of the former and, in the pertinent case, the polarization saturation of the latter. As an example to visually illustrate this point, Figs. 8.4 (a) and (b) show contour plots of the local invariants \(I_1(X)\) and \(I_5^E(X)\) from one of the FE solutions for the case of rigid high-permittivity particles with polarization saturation \(p_s = 10^{-4}\) C/m\(^2\) at volume fraction \(c = 0.05\).

An immediate observation from Fig. 8.3 is that the variational solution \((8.47)\) is in good quantitative agreement with corresponding FE solutions for all three types of filler particles considered. The variational solution \((8.47)\) is also seen to be in good qualitative agreement with the FE results in that it is independent of the invariants \(I_2\) and \(I_6^E\). This remarkable functional trait had already been observed in the simpler uncoupled context of the overall nonlinear elastic response of non-Gaussian rubber isotropically filled with rigid particles (Lopez-Pamies et al., 2013b), which corresponds to setting \(\bar{E} = 0\) and \(\mu_p = +\infty\) in the present context. A large body of results (in addition to those presented here) has confirmed that the variational solution \((8.47)\) remains in good qualitative and quantitative agreement with “exact” numerical solutions for finite deformations and finite electric fields irrespectively of the elastic dielectric properties of the dielectric elastomer and filler particles, as characterized by \(\Psi(I_1), \varepsilon, \mu_p, S(I_5^E)\), and irrespectively of the specifics of the underlying
microstructure, at least for volume fractions of particles sufficiently away from percolation.
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Figure 8.3: Plots of the effective free energy $\mathcal{W}$ for a non-Gaussian silicone elastomer, characterized by the free-energy function (8.110) with the material parameters listed in Table 8.1, filled with: (a)–(b) a volume fraction $c = 0.05$ of rigid conducting spherical particles, (c)–(d) a volume fraction $c = 0.05$ of rigid high-permittivity spherical particles with polarization saturation $p_s = 10^{-4}$ C/m$^2$, and (e)–(f) a volume fraction $c = 0.15$ of liquid-like high-permittivity spherical particles. Results are shown for the values of the free energy in terms of the invariants $I_2$ and $T_6^E$ for two sets of fixed values of the remaining four isotropic invariants. The solid lines (labeled “Sph. Theory”) correspond to appropriate specializations of the variational solution (8.47), namely, expressions (8.81), (8.78), and (8.84) for rigid conducting, rigid with polarization-saturating, and liquid-like high-permittivity monodisperse spherical particles, respectively. The dashed lines (labeled “Sph. FE”) correspond to the FE solutions.
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Figure 8.4: Representative contour plots of the local invariants $I_1(X)$ and $I_5^E(X)$ in a non-Gaussian silicone elastomer, characterized by the free energy function (8.110) with the material parameters listed in Table 8.1, filled with a volume fraction $c = 0.05$ of rigid high-permittivity spherical particles with polarization saturation. The plots correspond to values of the macroscopic invariants $I_1 = 3.74$ and $I_5^E = 49.8 \times 10^3 \text{ MV}^2/\text{m}^2$.

8.4.2 Rigid conducting particles

Figure 8.5 presents results determined from the variational solution (8.47) for the macroscopic response of the non-Gaussian silicone elastomer, again, characterized by the free-energy function (8.110) with the material parameters listed in Table 8.1, filled with rigid conducting particles under the conditions of electrostriction (8.88). Parts (a) and (c) of the figure pertain to results (labeled “HJ Theory”) for the iterative microstructure described in Section 8.2.1, wherein the particles are infinitely polydisperse in size. On the other hand, parts (b) and (d) correspond to the case of monodisperse spherical particles (labeled “Sph. Theory”) described in Section 8.2.2. Parts (e) and (f) include results for both microstructures. To further illustrate the accuracy of the variational solution (8.47), the FE solutions for the microstructure with spherical particles are also included in the appropriate parts of the figure (up to the point at which we were able to compute them). These solutions are labeled “Sph. FE” and are displayed as dashed lines or solid circles. The response of the unfilled silicone elastomer (dotted line) is also displayed in the figures for comparison purposes.

The plots in Figs. 8.5(a) through (d) show results for the electrostriction stretch $\lambda$ in terms of the applied electric field $E$, as defined by equation (8.90), for two volume fractions of particles, $c = 0.05$ and $c = 0.15$. A plain observation from these four sets of plots is that, irrespectively of the microstructure, the addition of rigid conducting particles has little effect on the electrostriction response of the silicone elastomer, at least up to around the stretch $\lambda \approx 0.62$ corresponding to the point at which the electric field $E$ reaches a local maximum, $E \approx 74.2 \text{ MV/m}$, in the unfilled silicone elastomer. Indeed, up to that point, the dielectric elastomer composite with the iterative microstructure deforms only slightly less than the unfilled silicone elastomer for both volume fractions.
of particles considered, while the dielectric elastomer composite with spherical particles deforms only slightly more. For sufficiently large electric fields $E > 74.2$ MV/m, the presence of rigid conducting particles does consistently lead to a sizable reduction in the electrostriction when compared to that of the unfilled silicone elastomer. In particular, larger volume fractions $c$ of particles lead to larger reductions. These results are in striking disagreement with most experimental investigations, which have reported enhancements in electrostriction up to several thousands of a percent for small additions ($c < 0.1$) of stiff conducting particles (see, e.g., Huang et al., 2005). The implications of this disagreement are discussed in Section 8.5.
Figure 8.5: Electrostriction response determined from the effective free energy (8.47) under conditions (8.88) — labeled “Theory” and displayed as solid lines in the plots — for a non-Gaussian silicone elastomer, characterized by the free energy function (8.110) with the material parameters listed in Table 8.1, filled with rigid conducting particles at volume fraction $c$. Results are shown for the infinitely polydisperse iterative microstructure (labeled “HJ”) and for the microstructure with monodisperse spherical particles (labeled “Sph.”). To further illustrate the accuracy of the variational solution (8.47), corresponding plots are also included of the FE solutions for the microstructure with spherical particles (labeled “FE” and displayed as dashed lines or solid circles).
Another salient observation from Figs. 8.5(a) through (d) is that, much like for the unfilled silicone elastomer, the applied electric field $E$ for the dielectric elastomer composites with volume fraction of particles $c = 0.05$ — but not for those with the larger volume fraction $c = 0.15$ — reaches a local maximum, $E_{LPD}$ say, at some critical electrostriction stretch, $\lambda_{LPD}$ say. As mentioned in remark viii above in Section 8.2 and discussed in detail in Section 5.2 of Part I, this point corresponds to the loss of positive definiteness of the tangent modulus of the effective Helmholtz free energy (8.43). The values of the critical electrostriction stretch $\lambda_{LPD}$ and corresponding critical electric field $E_{LPD}$ are plotted in Figs. 8.5(e) and (f) for both microstructures as functions of the volume fraction $c$ of particles. Remarkably, these plots indicate that the addition of rigid conducting particles has little effect on $\lambda_{LPD}$ and $E_{LPD}$ up to some threshold in $c$ beyond which any further addition of particles results in stable dielectric elastomers composites that remain positive definite for arbitrarily large electric fields $E$, in spite of the fact, again, that the underlying silicone matrix loses positive definiteness at $\lambda \approx 0.62$ and $E_{LPD} \approx 74.2$ MV/m. The threshold for the iterative microstructure with infinitely polydisperse particles is given approximately by $c = 0.095$, while that for the microstructure with monodisperse spherical particles is given approximately by $c = 0.089$.

### 8.4.3 Rigid high-permittivity particles with polarization saturation

Figures 8.6(a) through (d) show results analogous to those shown in Figs. 8.5(a) through (d) for the case of rigid high-permittivity particles with polarization saturation, whose behaviors are characterized by the free-energy function (8.111) with the material parameters indicated in Table 8.2. A quick glance at the plots suffices to recognize that the addition of particles with the larger polarization saturation $p_s = 10^{-2} \text{ C/m}^2$ leads to electrostriction responses that are not much different from the responses of the above-discussed dielectric elastomer composites wherein the particles are electrically conducting, especially for the case of volume fraction of particles $c = 0.05$. On the other hand, the addition of particles with the smaller polarization saturation $p_s = 10^{-4} \text{ C/m}^2$ consistently leads to a drastic reduction in the electrostriction response for all values of the applied electric field $E$ when compared to the response of the unfilled silicone elastomer. These results are, again, in disagreement with a number of experimental investigations, which have reported significant enhancements in the electrostriction response of dielectric elastomers when filled with small amounts ($c < 0.1$) of stiff polarization-saturating particles (see, e.g., Liu et al., 2013). The implications of this disagreement too are discussed in Section 8.5.
To gain further insight into the effects of the polarization saturation properties of the filler particles, Figs. 8.6(e) and (f) present results, as functions of $p_s$, for the critical electrostriction stretch $\lambda_{LPD}$ and associated critical electric field $E_{LPD}$ at which the dielectric elastomer composites with polarization-saturating particles reach an electromechanical limit load. Results are shown for both microstructures, with infinitely polydisperse particles (“HJ”) and with monodisperse spherical particles (“Sph.”), for a volume fraction of particles $c = 0.05$. It is evident from the plots that the electromechanical limit load in these classes of dielectric elastomer composites is largely unaffected by the polarization saturation of the particles.

A final point worth remarking from all six sets of plots included in Fig. 8.6 is that both microstructures (“HJ” and “Sph.”) exhibit nearly identical behaviors. This suggests that the response of non-Gaussian dielectric elastomers isotropically filled with rigid high-permittivity particles with polarization saturation is fairly insensitive, sufficiently away from percolation, to fine microstructural details (such as the size dispersion and the shape of the filler particles) beyond the volume fraction of particles, even more so than the non-Gaussian dielectric elastomers isotropically filled with rigid conducting particles discussed in the preceding subsection.
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Figure 8.6: Electrostriction response determined from the effective free energy (8.47) under conditions (8.88) — labeled “Theory” and displayed as solid lines in the plots — for a non-Gaussian silicone elastomer, characterized by the free energy function (8.110) with the material parameters listed in Table 8.1, filled with rigid high-permittivity particles with polarization saturation at volume fraction $c$. The behavior of the particles is characterized by the free-energy function (8.111) with the material parameters indicated in Table 8.2. Results are shown for the infinitely polydisperse iterative microstructure (labeled “HJ”) and for the microstructure with monodisperse spherical particles (labeled “Sph.”). Corresponding FE solutions are also included for the microstructure with spherical particles (labeled “FE” and displayed as dashed lines).
8.4.4 Liquid-like high-permittivity particles

Finally, we consider the electrostriction response of the non-Gaussian silicone elastomer when filled with liquid-like high-permittivity particles, whose behaviors are characterized by the free-energy function (8.111) with the material parameters indicated in Table 8.2. Results analogous to those presented in Fig. 8.5 are shown in Fig. 8.7. As opposed to the addition of rigid conducting particles and of rigid high-permittivity particles with polarization saturation, the addition of liquid-like high-permittivity particles is seen to lead to significant enhancements in electrostriction. In particular, larger volume fractions of particles consistently lead to larger enhancements. This is accompanied, however, by a sizable monotonic decrease in the limiting electric field $E_{LPD}$ while the corresponding stretch $\lambda_{LPD}$ remains fairly constant. The authors are not aware of any experimental investigation on dielectric elastomers isotropically filled with liquid-like high-permittivity particles. The results presented in Fig. 8.7 certainly motivate their pursuit.

We conclude by remarking that similar to the two previous cases involving rigid filler particles, all four sets of plots displayed in Fig. 8.7 show that both microstructures (“HJ” and “Sph.”) exhibit nearly identical behaviors. This suggests that the response of non-Gaussian dielectric elastomers isotropically filled with liquid-like particles too is largely insensitive to fine microstructural details beyond the volume fraction of particles (again, sufficiently away from percolation). Further evidence supporting this lack of sensitivity is provided by the fact that, up to the volume fraction of particles considered in this chapter $c = 0.25$, the FE solutions for the isotropic distribution of polydisperse spherical particles with three different sizes described in Section 8.3 are virtually indistinguishable from those presented in Figs. 8.5 through 8.7 for monodisperse particles.
8. Nonlinear electroelastic deformations of DECs: Non-Gaussian elastic dielectrics

Figure 8.7: Electrostriction response determined from the effective free energy (8.47) under conditions (8.88) — labeled “Theory” and displayed as solid lines in the plots — for a non-Gaussian silicone elastomer, characterized by the free energy function (8.110) with the material parameters listed in Table 8.1, filled with liquid-like high-permittivity particles at volume fraction \( c \). The behavior of the particles is characterized by the free-energy function (8.111) with the material parameters indicated in Table 8.2. Results are shown for the infinitely polydisperse iterative microstructure (labeled “HJ”) and for the microstructure with monodisperse spherical particles (labeled “Sph.”). Corresponding FE solutions are also included for the microstructure with spherical particles (labeled “FE” and displayed as dashed lines).
8.5 Comparisons with experimental data

Most experimental investigations have reported that dielectric elastomers filled with small amounts of (semi-)conductive or high-permittivity particles exhibit electrostriction properties that are far superior to those of the corresponding unfilled dielectric elastomers (see, e.g., Zhang et al., 2002; Huang and Zhang, 2004; Huang et al., 2005; Carpi and De Rossi, 2005; Mc Carthy et al., 2009; Meddeb and Ounaies, 2012; Liu et al., 2013). Motivated by a heuristic analysis in the asymptotic context of small deformations and moderate electric fields, Li (2003) and Li et al. (2004) conjectured that such superior properties are due to the nonlinear elastic dielectric nature of elastomers which heightens the role of the fluctuations of the electric field in the presence of filler particles. Still within the asymptotic context of small deformations and moderate electric fields, Tian et al. (2012) extended and made rigorous the approximate analysis of Li (2003) and Li et al. (2004) and also pointed to the nonlinear elastic dielectric nature of elastomers as the dominant electrostriction enhancing mechanism. Armed with the general analytical solution (8.47) and the hybrid FE formulation presented in Section 8.3, we are now in a position to probe the verity of this initial conjecture. Contrary to it, the sample results for finite deformations and finite electric fields presented in subsections 8.4.2 and 8.4.3 have already suggested that the nonlinear elastic dielectric nature of elastomers is not the mechanism responsible for the enhanced electrostriction properties exhibited by emerging dielectric elastomer composites. In the sequel, we directly confront the variational solution (8.47) and FE solutions to two sets of representative experiments that confirm that this is in fact the case.

We begin by examining the experimental data of Huang et al. (2005) for the electrostriction response under a uniaxial electric field of a polyurethane elastomer filled with semi-conducting o-CuPc particles. The particles were reported to be roughly spherical in shape, about 40 nm in average diameter, and, with help of a polyacrylic acid (PAA) coating, well dispersed with overall isotropic symmetry. The total initial volume fraction of the o-CuPc particles and surrounding PAA coating was reported to be $c = 0.073$. Figure 8.8(a) shows the measured electrostriction strain $\lambda - 1$ as a function of the applied electric field $E$ for the filled polyurethane elastomer (solid triangles); see Section 8.2.3 for the definition of these variables. To aid the discussion, Fig. 8.8(a) also shows the measured electrostriction response for the unfilled polyurethane elastomer (empty triangles).

In computing the theoretical response predicted by the effective free-energy function (8.47), given the partial information available, we take the polyurethane elastomer in the composite to be characterized by the free-energy function (8.110) with parameters $\mu_1 = 0.410$ MPa, $\mu_2 = 0.409$ MPa,
8. Nonlinear electroelastic deformations of DECs: Non-Gaussian elastic dielectrics

Figure 8.8: Comparisons between the theoretical predictions determined from the effective free-energy function (8.47) and the experimental data of Huang et al. (2005) and Liu et al. (2013) for the electrostriction of: (a) a polyurethane elastomer filled with semi-conducting o-CuPc particles at volume fraction $c = 0.073$ and (b) a silicone elastomer filled with polarization-saturating TiO$_2$ particles at volume fraction $c = 0.082$, under the application of a uniaxial electric field of magnitude $E$. The experimental (theoretical) data for the dielectric elastomer composites is depicted as solid triangles (solid lines), while the experimental (theoretical) data for the underlying unfilled elastomers is depicted as hollow triangles (dotted lines). For further scrutiny, plots are also included (dashed lines) of corresponding FE predictions for the dielectric elastomer composites.

$\alpha_1 = -8.034$, $\alpha_2 = 0.841$, and $\varepsilon = 8.0\varepsilon_0$, as fitted to the electrostriction and dielectric data provided by Huang et al. (2005) for the unfilled polyurethane elastomer$^3$. Note that this assumes that the synthesis process in the presence of the o-CuPc nanoparticles does not alter the properties of the resulting polyurethane. We further assume that the particles are spherical in shape and monodisperse in size; recall from Section 8.4 that at the small volume fraction of particles $c = 0.073$ of interest here, particle size dispersion has essentially no effect on macroscopic elastic dielectric properties. Moreover, since o-CuPc is a semi-conducting polymer with an initial shear modulus of about 1 GPa, we model the behavior of the particles by means of the free-energy function (8.111) with material parameters $\mu_p = 1$ GPa, $\varepsilon_p = +\infty$, and $p_s = +\infty$. The electrostriction response that results from the effective free-energy function (8.47) given these inputs is displayed in Fig. 8.8(a) as a solid line. The corresponding FE prediction is also displayed in the figure as a dashed line. As expected from the sample results of Section 8.4.2, the theoretical results show little difference between the electrostriction of the dielectric elastomer composite and that of the unfilled polyurethane. This is in striking disagreement with the experimental data, which shows that the electrostriction of the dielectric elastomer composite is about 20 times larger than that of the unfilled polyurethane.

$^3$It is of note that the electrostriction data of Huang et al. (2005) imply a nonlinear elastic response of polyurethane that is about an order of magnitude softer than other experimental investigations have reported (see, e.g., Qi and Boyce, 2005).
We now turn to examine the experimental data of Liu et al. (2013) for the electrostriction response under a uniaxial electric field of a silicone elastomer isotropically filled with roughly spherical TiO$_2$ particles of about 3 $\mu$m in average diameter at volume fraction $c = 0.082$. Figure 8.8(b) shows the reported transverse (or so-called actuation) electrostriction $\lambda^{-1/2}$ as a function of the applied electric field $E$ for the filled silicone elastomer (solid triangles). Figure 8.8(b) also includes the electrostriction response for the unfilled silicone elastomer (empty triangles).

At the level of the variational solution (8.47), much like in the previous comparison, we take the silicone elastomer in the composite to be characterized by the free-energy function (8.110) with parameters $\mu_1 = 0.032$ MPa, $\mu_2 = 0.023$ MPa, $\alpha_1 = 3.837$, $\alpha_2 = 0.559$, and $\varepsilon = 3.2\varepsilon_0$, as fitted to the reported electrostriction and dielectric data for the unfilled silicone elastomer. Moreover, the particles are assumed to be spherical in shape and monodisperse in size. Given their polycrystalline rutile composition, we take their elastic dielectric behavior to be characterized by the free-energy function (8.111) with initial shear modulus $\mu_p = 110$ GPa, initial permittivity $\varepsilon_p = 114\varepsilon_0$, and polarization saturation $p_s = 10^{-2}$ C/m$^2$. The electrostriction response that results from the effective free-energy function (8.47) given these inputs is displayed in Fig. 8.8(b) as a solid line. For further scrutiny of the experimental data, the corresponding FE solution is also included (dashed line) in the figure. Again, as expected from the sample results of Section 8.4.3, the theoretical predictions for the electrostriction response of the dielectric elastomer composite are practically indistinguishable from the response of the unfilled silicone elastomer. In stark contrast, the experimental data for the dielectric elastomer composite shows about a 50% enhancement in electrostriction with respect to the unfilled silicone.

The above glaring disagreement between the theoretical results and experiments indicates that the basic point of view (adopted throughout this work until this point) that dielectric elastomer composites are two-phase particulate elastic dielectric composites is fundamentally incomplete, especially for cases involving stiff filler particles such as those shown in Fig. 8.8. In this regard, we recall that in elastomers filled with stiff particles the “anchoring” of the underlying polymer chains to the filler particles forces the chains into conformations that are very different from those in the bulk and that this results in “interphases” of different mechanical and physical behavior. The presence of such interphases (often referred to as bound rubber in the rubber science community) has long been known to have major effects on macroscopic properties when the filler particles are
submicron in size (see, e.g., Leblanc, 2010; Goudarzi et al., 2015 and references therein). Furthermore, space charges in such interphases may be present from the outset because of the fabrication process of the materials (see, e.g., Bauer et al., 2004; Lewis, 2004; Roy et al., 2005; Deng et al., 2014). They may also be injected from the particles upon the application of an electric field (see, e.g., Lewis, 2004; Roy et al., 2005). Whatever their origin, the presence of space interphasial charges has been recently shown to have the potential to lead to extreme enhancements of the macroscopic dielectric response of particulate composites and, by the same token, extreme enhancements of their electrostrictive response (Lopez-Pamies et al., 2014). We posit that the extreme enhancements in electromechanical properties that have been exhibited by emerging dielectric elastomer composites are the manifestation of the above-described interphasial phenomena.

![Figure 8.9](image_url)

Figure 8.9: (a) Comparison between the experimental electrostriction data (solid triangles) of Huang et al. (2005) for a polyurethane elastomer filled with an isotropic distribution of semi-conducting o-CuPc particles at volume fraction $c = 0.073$ and a theoretical prediction (solid line) featuring interphasial charges. In particular, the latter corresponds to the FE solution for the electrostriction response of the elastic dielectric composite whose defining unit cell is depicted in (b), namely, a non-Gaussian dielectric elastomer (with free-energy function(8.110) and material parameters $\mu_1 = 0.410$ MPa, $\mu_2 = 0.409$ MPa, $\alpha_1 = -8.034$, $\alpha_2 = 0.841$, $\varepsilon = 8.0\varepsilon_0$) filled with an isotropic distribution of rigid conducting monodisperse spherical particles at volume fraction $c = 0.073$ that are surrounded by the distribution of space charges described by expression (8.112) with $R_m = 20$ nm, $t = 3$ nm, and $q_i = 6500\varepsilon_0$. The FE solution for the same elastic dielectric composite but without the interphasial charges (dashed line), already displayed in Fig. 8.8(a), is also included in this figure for comparison purposes.

While it is known that the elastomeric matrix material in a given filled elastomer may not have the same mechanical and physical properties as the “same” elastomer synthesized in the absence of filler particles, the expected range of possible differences cannot account for the extreme enhancements observed experimentally. Viscous and dielectric dissipative phenomena might also contribute to the observed enhancements. Preliminary calculations in the context of time-dependent dielectric composite materials have suggested, however, that they are of lesser importance than interphasial phenomena (Ladeb and Lopez-Pamies, 2015).
By way of an example, in support of the above conjecture, Fig. 8.9(a) illustrates the close agreement possible between a theoretical prediction incorporating interphasial charges and the above-discussed experimental data of Huang et al. (2005) for a polyurethane elastomer filled with semiconducting o-CuPc particles at volume fraction $c = 0.073$. The theoretical prediction corresponds to the FE solution for the electrostriction response of a non-Gaussian dielectric elastomer filled with an isotropic distribution of monodisperse spherical particles that are surrounded by a layer of space charges. Specifically, like in the theoretical results presented in Fig. 8.8(a), the elastic dielectric behavior of the non-Gaussian dielectric elastomer is taken to be characterized by the free-energy function (8.110) with parameters $\mu_1 = 0.410$ MPa, $\mu_2 = 0.409$ MPa, $\alpha_1 = -8.034$, $\alpha_2 = 0.841$, and $\varepsilon = 8.0\varepsilon_0$, as fitted to the electrostriction and dielectric data provided by Huang et al. (2005) for the unfilled polyurethane elastomer. The filler particles are taken to be rigid and electrically conducting.

Finally, following Lopez-Pamies et al. (2014), the distribution of space charges that surround the particles is taken to be characterized by the charge density (per unit undeformed volume)

$$Q(X) = \theta_i(X)q_i \frac{E \cdot (X - X^c)}{R_m |X - X^c|} \quad \text{with} \quad \theta_i(X) = \begin{cases} 1 & \text{if } R_m < |X - X^c| < R_m + t \\ 0 & \text{otherwise} \end{cases}.$$  

(8.112)

Here, $R_m$ and $X^c$ stand for the common radius and the position vector of the center of each spherical particle, $t$ denotes the common constant thickness of the spatial regions around each particle where the charges are contained, and $q_i$ is a constant of choice (of units F/m) that physically can be viewed as a measure of charge content. The FE result presented in Fig. 8.9(a) corresponds to the values $R_m = 20$ nm, $t = 3$ nm, and $q_i = 6500\varepsilon_0$. Figure 8.9(b) illustrates the unit cell whose periodic repetition defines the precise microstructure in this example.

We emphasize that expression (8.112) is an ad hoc constitutive choice and that little is actually known about interphasial charges in dielectric elastomer composites. Further studies in this direction appear to be of the essence in order to understand the fundamental microscopic mechanisms behind the remarkable electromechanical properties that dielectric elastomer composites seem capable of achieving.
Homogenization of elastic dielectric composites with rapidly oscillating passive and active source terms

I want to be reminded and delighted and surprised once again, through interplanetary exploration, with the infinite variety and novelty of phenomena that can be generated from such simple principles.

– Richard P. Feynman, The Feynman Lectures on Physics, 1964

Motivated by the sample results for the remarkable electrostrictive properties of dielectric elastomer composites comprising interphasial charges presented in the previous chapter (see Fig. 8.9(a)), we carry out in this chapter a formal derivation for the homogenized equations governing the macroscopic response of elastic dielectric composites that contain space charges that oscillate rapidly in space, in the so-called limit of small deformations and moderate electric fields. The focus is on elastic dielectric composites with even electromechanical coupling and periodic microstructure, which contain rapidly oscillating space charges of two types: passive and active.

Passive space charges refer to space charges that are present within the elastic dielectric composite from the outset, in its ground state. A prominent class of materials that can be viewed as elastic dielectric composites containing passive space charges is electrets (see, e.g., Hilczer and Malecki, 1986; Gerhard-Multhaupt, 1999; Kestelman et al., 2000; Bauer et al., 2004; Hillenbrand and Sessler, 2008; Deng et al., 2014). On the other hand, active space charges refer to space charges that are not present within the elastic dielectric composite in its ground state. Instead, they appear within
the composite as a result of externally applied stimuli, for instance, by a charge injection process (Lewis, 2004; Roy et al., 2005). Dielectric elastomers filled with (semi-)conducting or high-dielectric nanoparticles are thought to be an example of such a class of materials (Lewis, 2004; Lopez-Pamies et al., 2014; see also Section 8.5 in Chapter 8). In this chapter, we shall consider active charges that appear in proportionality to the electric field induced within the composite by externally applied electrical stimuli.

9.1 The problem

*Ergo*, consider an elastic dielectric composite with periodic microstructure of period $\delta$ that occupies a bounded domain $\Omega \subset \mathbb{R}^N$ ($N = 1, 2, 3$), with smooth boundary $\partial \Omega$ and closure $\overline{\Omega} = \Omega \cup \partial \Omega$, in its undeformed configuration; throughout this work, again, attention is restricted to elastic dielectrics with even electromechanical coupling. In the classical setting of small deformations and moderate electric fields (see the Appendix), the permittivity, elasticity, and electrostriction tensors that characterize the local elastic dielectric response of the composite at any material point $X \in \Omega$ are taken to be, without loss of generality and with help of the notation $Y = (0, 1)^N$, of the form

$$
\varepsilon_\delta^{ij}(X) \in \mathbb{R}, \quad \varepsilon_\delta^{ij}(X) = \varepsilon_{ij}(\delta^{-1}X) \quad \text{with} \quad \varepsilon_{ij}(y) \text{--periodic},
$$

$$
L_\delta^{ijkl}(X) \in \mathbb{R}, \quad L_\delta^{ijkl}(X) = L_{ijkl}(\delta^{-1}X) \quad \text{with} \quad L_{ijkl}(y) \text{--periodic},
$$

$$
M_\delta^{ijkl}(X) \in \mathbb{R}, \quad M_\delta^{ijkl}(X) = M_{ijkl}(\delta^{-1}X) \quad \text{with} \quad M_{ijkl}(y) \text{--periodic},
$$

respectively. Basic physical considerations dictate that

$$
\varepsilon_\delta^{ij} = \varepsilon_\delta^{ji}, \quad \varepsilon_\delta^{ij} \xi_i \xi_j \geq \varepsilon_0 \xi_k \xi_k \quad \forall \xi \in \mathbb{R}^N,
$$

$$
L_\delta^{ijkl} = L_\delta^{klij} = L_\delta^{jikl} = L_\delta^{ijlk}, \quad L_\delta^{ijkl} \Xi_{ij} \Xi_{kl} \Xi_{pq} \Xi_{pq} \quad \forall \Xi \in \mathbb{R}^{N \times N},
$$

$$
M_\delta^{ijkl} = M_\delta^{jikl} = M_\delta^{ijlk},
$$

where $\varepsilon_0 \approx 8.85 \times 10^{-12} \text{ F/m}$ stands for the permittivity of vacuum and $\theta$ is some positive constant, namely, the smallest eigenvalue of $L_\delta^{ijkl}$, which is required to be positive. For mathematical expediency, we assume the following regularity properties:

$$
\varepsilon_\delta^{ij} \in C^1(\overline{\Omega}), \quad L_\delta^{ijkl} \in C^\infty(\Omega), \quad M_\delta^{ijkl} \in C^\infty(\Omega).
$$

Here, we remark that the relatively strong regularity (9.5) of the components of the permittivity tensor $\varepsilon^\delta(X)$ is invoked in order to leverage standard theorems (in particular, the Lax-Milgram
9. Homogenization of elastic dielectric composites with rapidly oscillating source terms

Theorem) that will warrant mathematical well posedness; more precisely, as elaborated below, the regularity (9.5) is invoked here in order to obtain the sufficient regularity for the electric fields needed to prove existence of solution for the mechanical fields via the Lax-Milgram theorem. Such a regularity can be relaxed to allow for a general class of piecewise constant values of $\varepsilon^\delta(X)$ — for instance, the piecewise constant values of $\varepsilon^\delta(X)$ associated with particulate composites wherein the inclusions have smooth boundaries — at the expense of possibly invoking more technical theorems (see, e.g., Avellaneda and Lin, 1987; Li and Nirenberg, 2003). We assume further that the composite is subjected to a prescribed electric potential and a prescribed displacement

$$\phi \in H^{3/2}(\partial\Omega) \quad \text{and} \quad v \in H^{1/2}(\partial\Omega; \mathbb{R}^N)$$

(9.6)
on the entirety of its boundary $\partial\Omega$; Neumann or mixed boundary conditions could be considered at no significant further conceptual expense. Moreover, we assume that the composite contains a distribution of space charges with density (per unit undeformed volume)

$$q^\delta \in L^2(\Omega).$$

(9.7)

Figure 9.1 illustrates a schematic of the composite and of its microstructure and space charge content.

---

Figure 9.1: (a) Schematic of the elastic dielectric composite in its undeformed configuration $\Omega$; the boundary layer of incomplete unit cells needed to conform with the arbitrary geometry of its boundary $\partial\Omega$ is marked in red. (b) Schematic of the unit cell $Y$ that defines the periodic microstructure (of period $\delta$) of the composite with the explicit illustration of the distribution of space charges characterized by the space-charge density $q^\delta(X)$.

In the limit of small deformations and moderate electric fields (see the Appendix), the relevant equations of Maxwell and of balance of linear momentum can be shown to reduce to the following
one-way coupled boundary-value problems:

\[
\begin{aligned}
\frac{\partial}{\partial X_i} \left[ -\varepsilon_{ij}^\delta(X) \frac{\partial \varphi^\delta(X)}{\partial X_j} \right] &= q^\delta(X), \quad X \in \Omega \\
\varphi^\delta(X) &= \phi(X), \quad X \in \partial \Omega
\end{aligned}
\] (9.8)

and

\[
\begin{aligned}
\frac{\partial}{\partial X_j} \left[ L_{ijkl}^\delta(X) \frac{\partial u^\delta_i(X)}{\partial X_k} + M_{ijkl}^\delta(X) \frac{\partial \varphi^\delta(X)}{\partial X_k} \frac{\partial \varphi^\delta(X)}{\partial X_l} \right] &= 0, \quad X \in \Omega \\
u^\delta_i(X) &= v_i(X), \quad X \in \partial \Omega
\end{aligned}
\] (9.9)

for the electric potential \( \varphi^\delta(X) \) and the displacement field \( u^\delta(X) \). The pde (9.8)\textsubscript{1} is the standard equation that governs the electrostatic field within a dielectric medium that contains a distribution of space charges. We remark that its restriction to the domain \( \Omega \) occupied by the solid (as opposed to the entire space \( \mathbb{R}^N \) where Maxwell’s equations ought to be solved) is sufficient in the present context thanks to the prescription of the Dirichlet boundary condition (9.8)\textsubscript{2}. On the other hand, the pde (9.9)\textsubscript{1} governs the deformation of the solid that results from the electric field in addition to the applied displacement boundary condition (9.9)\textsubscript{2}.

From a mathematical point of view, we remark that while the coupled system of boundary-value problems (9.8)–(9.9) is nonlinear, the boundary-value problem (9.8) is linear in the electric potential \( \varphi^\delta(X) \) and the boundary-value problem (9.9) is linear in the displacement field \( u^\delta(X) \). For any fixed \( \delta > 0 \) then, granted the ellipticity (9.2)\textsubscript{2} and regularity (9.5)\textsubscript{1} of the components of the permittivity tensor \( \varepsilon^\delta(X) \), the properties (9.6)\textsubscript{1} and (9.7) of the boundary data and source term, and the smoothness of \( \partial \Omega \), the Lax-Milgram theorem ensures existence and uniqueness of the solution of (9.8) for \( \varphi^\delta(X) \) in the Sobolev space \( H^1(\Omega) \). The regularity (9.5)\textsubscript{1}, (9.6)\textsubscript{1}, (9.7) together with the smoothness of \( \partial \Omega \) imply in fact the stronger regularity result that \( \varphi^\delta \in H^2(\Omega) \), and hence that \( \text{Grad} \varphi^\delta \in H^1(\Omega; \mathbb{R}^N) \subset L^4(\Omega; \mathbb{R}^N) \); see, e.g., Chapter 8 in Gilbarg and Trudinger (2001), Chapter 6.3 in Evans (2010), Theorem 8.3 in Lions and Megenes (1972), and Theorem 9.16 in Brezis (2011). In turn, granted the ellipticity (9.3)\textsubscript{2} and boundedness (9.5)\textsubscript{2} of the components of the elasticity tensor \( L^\delta(X) \), the boundedness (9.5)\textsubscript{3} of the components of the electrostriction tensor \( M^\delta(X) \), the fact that \( \text{Grad} \varphi^\delta \in L^4(\Omega; \mathbb{R}^N) \) so that by (the generalized) Hölder’s inequality \( \text{Grad} \varphi^\delta \otimes \text{Grad} \varphi^\delta \in L^2(\Omega; \mathbb{R}^{N \times N}) \), the regularity (9.6)\textsubscript{2} of the boundary data, and the smoothness of \( \partial \Omega \), the Lax-Milgram theorem ensures existence and uniqueness of the solution of (9.9) for the displacement field \( u^\delta(X) \) in the Sobolev space \( H^1(\Omega; \mathbb{R}^N) \).
A specific class of space-charge densities $q^\delta(X)$ We shall restrict attention here to space-charge densities $q^\delta(X)$ of the following divergence form:

$$q^\delta(X) = -\delta \frac{\partial}{\partial X_i} \left[ f_k(X) \frac{\partial}{\partial X_i} [\psi_k(\delta^{-1}X)] \right] = \delta^{-1} f_k(X) g_k(\delta^{-1}X) - \frac{\partial f_k}{\partial X_i}(X) \tau_{ki}(\delta^{-1}X).$$  

(9.10)

Here, $f \in H^2(\Omega; \mathbb{R}^N), \ g$ is $Y$-periodic, $g \in L^\infty(Y; \mathbb{R}^N), \ \int_Y g(y)dy = 0$ (9.11)

and $\tau_{ki}(y) = \frac{\partial \psi_k(y)}{\partial y_i}$ with $\psi(y)$ defined in terms of $g(y)$ as the unique solution in $H^2(Y; \mathbb{R}^N)$ of the linear elliptic boundary-value problem

$$\begin{align*}
-\frac{\partial^2 \psi_k}{\partial y_i \partial y_i}(y) = g_k(y), & \quad y \in Y \\
-\frac{\partial \psi_k}{\partial y_i}(y)n_i = 0, & \quad y \in \partial Y \\
\int_Y \psi_k(y)dy = 0
\end{align*}$$  

(9.12)

where $n$ in (9.12)$_2$ stands for the outward unit normal to the boundary $\partial Y$ of the unit cell $Y$ (see Fig. 9.1b).

The choice (9.10) with (9.11)$_{2,4}$ and (9.12) of space-charge density is motivated by physical requirements/observations as well as by mathematical expediency. Indeed, the divergence form (9.10) together with the zero-average condition (9.11)$_4$ and the boundary condition (9.12)$_2$ ensure global charge neutrality in $\Omega$ up to a boundary layer of thickness $\delta$ (see Fig. 1a). Moreover, the leading order term in (9.10) being $O(\delta^{-1})$ implies that the content of charges at the “microscopic” length scale $\delta$ remains finite even in the limit as $\delta \to 0$ (in this limit, the space-charge density $q^\delta(X)$ blows up within a vanishingly small volume to lead to a microscopic distribution of finite charges), consistent with physical expectations. We finally remark that the form (9.10) comprises two constitutive inputs: the functions $f(X)$ and $g(\delta^{-1}X)$. Roughly speaking, the latter dictates the local distribution of charges at the microscopic length scale $\delta$ of each unit cell. The former, on the other hand, dictates the possibly non-uniform distribution of charges at the macroscopic length scale of $\Omega$. Finally, it is also interesting to note that source terms of the asymptotic form (9.10) — with leading $O(\delta^{-1})$ and correction $O(\delta^0)$ — can appear naturally when converting elliptic boundary-value problems with non-homogeneous Dirichlet boundary conditions to problems with homogeneous Dirichlet boundary conditions; see, e.g. Section 18 of Chapter 1 in the monograph by Bensoussan et al. (1978).
9.2 Passive charges: the limit as $\delta \to 0$ by the method of two-scale asymptotic expansions

In this section, we present the derivation of the homogenized equations that emerge from the boundary-value problems (9.8)–(9.9) in the limit as $\delta \to 0$ by means of the method of two-scale asymptotic expansions (Sanchez-Palencia, 1980; Bensoussan et al., 1978). In the present context, this method amounts to looking for an asymptotic solution of the equations (9.8)–(9.9) as $\delta \to 0$ of the form

$$
\varphi^\delta(X) = \sum_{k=0}^{\infty} \delta^k \varphi^{(k)}(X, \delta^{-1}X) \quad \text{and} \quad u^\delta(X) = \sum_{k=0}^{\infty} \delta^k u^{(k)}(X, \delta^{-1}X),
$$

(9.13)

where the functions $\varphi^{(k)}(X, \delta^{-1}X)$ and $u^{(k)}(X, \delta^{-1}X)$ are $Y$-periodic in their second argument and, according to the boundary conditions (9.8)$_2$ and (9.9)$_2$, such that $\varphi^{(0)}(X, \delta^{-1}X) = \phi(X)$, $\varphi^{(k)}(X, \delta^{-1}X) = 0$ for $k \neq 0$, $u^{(0)}(X, \delta^{-1}X) = v(X)$, and $u^{(k)}(X, \delta^{-1}X) = 0$ for $k \neq 0$ on $\partial\Omega$. In view of the one-way coupling of the boundary-value problems (9.8)–(9.9), we begin in Section 9.2.1 by working out the limit for the electric potential $\varphi^\delta(X)$ and subsequently make use of this result to then work out the limit for the displacement field $u^\delta(X)$ in Section 9.2.2.

A few words about the presentation are in order. A number of the results that are obtained in Section 9.2.1 are classical, yet we opt to include their presentation in order to preserve the continuity of the derivation and, more critically, to better be able to point to how the presence of space charges affects the homogenized equations. Similarly, some of the results that are obtained in Section 9.2.2 have been previously obtained by Tian (2007) (see also Tian et al., 2012) via the two-scale convergence method (Allaire, 1992). In addition to providing an alternative derivation for those, their inclusion in the presentation here preserves the continuity of the derivation and, more critically, aids in illustrating how the addition of space charges (not present in the work of Tian (2007) impacts the homogenized equations.

Before proceeding with the derivation per se, it is important to remark that while the method of two-scale asymptotic expansions typically yields the right homogenized equations (see, e.g., Bensoussan et al., 1978), it is not a rigorous proof of the homogenization limit; this is because the two-scale ansatz, (9.13) for the problem of interest here, may possibly be incorrect beyond $O(\delta)$ due, for instance, to boundary-layer effects in the vicinity of $\partial\Omega$; see, e.g., Sanchez-Palencia, (1987);...
Allaire and Amar, (1999) and references therein. The rigorous proof that the homogenized equations derived here from the two-scale asymptotic expansion are indeed correct turns out to be quite technical because of the quadratic term Grad $\varphi^\delta(X) \otimes \text{Grad} \varphi^\delta(X)$ in equation (9.9) for the balance of linear momentum. Such a rigorous proof will be presented elsewhere.

9.2.1 The limit of the electric potential $\varphi^\delta(X)$ as $\delta \rightarrow 0$

Upon introducing the variables $x = X$ and $y = \delta^{-1}X$ and the operator

$$\mathbf{A}^\delta = \delta^{-2} A^{(1)} + \delta^{-1} A^{(2)} + \delta^0 A^{(3)}$$

with

$$A^{(1)} = -\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(y) \frac{\partial}{\partial y_j} \right],$$

$$A^{(2)} = -\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(y) \frac{\partial}{\partial x_j} \right] - \frac{\partial}{\partial x_i} \left[ \varepsilon_{ij}(y) \frac{\partial}{\partial y_j} \right],$$

$$A^{(3)} = -\frac{\partial}{\partial x_i} \left[ \varepsilon_{ij}(y) \frac{\partial}{\partial x_j} \right],$$

where $\frac{\partial}{\partial x_i}$ and $\frac{\partial}{\partial y_i}$ denote partial derivatives with respect to $x$ and $y$, we begin by recasting the pde (9.8) for the electric potential $\varphi^\delta(X)$ in the more convenient form

$$A^\delta \varphi^\delta = \eta^\delta.$$  \hspace{1cm} (9.16)

Substituting the ansatz (9.13) in the pde (9.16) and expanding in powers of $\delta$ leads to a hierarchy of equations of a very distinctive structure for the functions $\varphi^{(k)}(x,y)$. The first three of these equations turn out to be enough for our purposes here, namely, to determine the first two terms $\varphi^{(0)}(x,y)$ and $\varphi^{(1)}(x,y)$ in the expansion (9.13). They are of $O(\delta^{-2})$, $O(\delta^{-1})$, $O(\delta^0)$ and in terms of the operators (9.15) read as

$$A^{(1)} \varphi^{(0)} = 0,$$

$$A^{(1)} \varphi^{(1)} + A^{(2)} \varphi^{(0)} = f_k(x) g_k(y),$$

$$A^{(1)} \varphi^{(2)} + A^{(2)} \varphi^{(1)} + A^{(3)} \varphi^{(0)} = -\frac{\partial f_k}{\partial x_i} (x) \frac{\partial \psi_k}{\partial y_i}(y).$$

The equation of order $\delta^{-2}$ The equation (9.17) of leading order is a pde for the function $\varphi^{(0)}(x,y)$ where $y$ is the independent variable and $x$ plays the role of a parameter. Its unique
solution (with respect to $y$) is simply a function of $x$ that does not depend on $y$. We write

$$
\varphi^{(0)}(x, y) = \varphi(x).
$$

(9.20)

The equation of order $\delta^{-1}$ Making direct use of relation (9.20), the equation (9.18) of order $\delta^{-1}$ reduces to

$$
- \frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(y) \frac{\partial \varphi^{(1)}(x, y)}{\partial y_j} \right] = \frac{\partial \varepsilon_{ij}(y)}{\partial y_i} \frac{\partial \varphi}{\partial x_j}(x) + f_k(x)g_k(y), \quad y \in Y,
$$

(9.21)

which, for a given function $\varphi(x)$ and a given $x$, can be thought of as a pde for the function $\varphi^{(1)}(x, y)$ in the periodic unit cell $Y$ with $x$ playing the role of a parameter. By introducing the $Y$-periodic functions $\omega_i(y)$ and $\varpi_i(y)$ defined implicitly as the unique solutions of the linear elliptic pdes

$$
\begin{cases}
\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(y) \frac{\partial \omega_k}{\partial y_j}(y) \right] = \frac{\partial \varepsilon_{ik}}{\partial y_i}(y), \quad y \in Y \\
\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(y) \frac{\partial \varpi_k}{\partial y_j}(y) \right] = g_k(y), \quad y \in Y \\
\int_Y \omega_k(y) dy = 0 \\
\int_Y \varpi_k(y) dy = 0
\end{cases}
$$

(9.22)

the unique solution (with respect to $y$) of (9.21) can be written as

$$
\varphi^{(1)}(x, y) = -\omega_k(y) \frac{\partial \varphi}{\partial x_k}(x) - \varpi_k(y)f_k(x) + r^{(1)}(x),
$$

(9.23)

where $r^{(1)}(x)$ is an arbitrary function of $x$.

The equation of order $\delta^0$ Making again direct use of relation (9.20), the equation (9.19) of order $\delta^0$ can be simplified to

$$
- \frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(y) \frac{\partial \varphi^{(2)}(x, y)}{\partial y_j} \right] = \frac{\partial \varepsilon_{ij}(y)}{\partial y_i} \frac{\partial \varphi^{(1)}(x, y)}{\partial x_j} + \\
\frac{\partial}{\partial x_i} \left[ \varepsilon_{ij}(y) \left( \frac{\partial \varphi}{\partial x_j}(x) + \frac{\partial \varphi^{(1)}(x, y)}{\partial y_j} \right) \right] - \frac{\partial f_k}{\partial x_i}(x) \frac{\partial \psi_k}{\partial y_i}(y), \quad y \in Y.
$$

(9.24)

For a given function $\varphi(x)$ and a given $x$ (since $\varphi^{(1)}(x, y)$ is given by (9.23) in terms of $\varphi(x)$), this equation can be thought of as a pde for the function $\varphi^{(2)}(x, y)$ in the periodic unit cell $Y$ with $x$ playing the role of a parameter.

Now, the pde (9.24) admits a solution (with respect to $y$ and unique up to an additive constant) for $\varphi^{(2)}(x, y)$ if its right-hand side has zero average over $Y$; this is the so-called Fredholm alternative. Consequently, after some manipulation employing the divergence theorem together with the $Y$-periodicity of $\varepsilon(y)$ and $\varphi^{(1)}(x, y)$, we require that

$$
\frac{\partial}{\partial x_i} \int_Y \left[ \varepsilon_{ij}(y) \left( \frac{\partial \varphi}{\partial x_j}(x) + \frac{\partial \varphi^{(1)}(x, y)}{\partial y_j} \right) - f_k(x) \frac{\partial \psi_k}{\partial y_i}(y) \right] dy = 0.
$$

(9.25)
Making use of the representation (9.23) for $\varphi^{(1)}(x,y)$ in terms of the $Y$-periodic functions $\omega_i(y)$ and $\varpi_i(y)$, the divergence theorem repeatedly, and exploiting the $Y$-periodicity of the pdes (9.22), this equation can be simplified to

$$\frac{\partial}{\partial x_i} \left[-\hat{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(x)\right] = \hat{q}(x), \quad (9.26)$$

where

$$\hat{\varepsilon}_{ij} = \int_Y \varepsilon_{ik}(y) \left( \delta_{jk} - \frac{\partial \omega_i}{\partial y_k}(y) \right) dy$$

and

$$\hat{q}(x) = -\frac{\partial}{\partial x_i} \left[ \hat{\alpha}_{ij} f_j(x) \right] \quad (9.27)$$

Equation (9.26) is the homogenized pde in $\Omega$ that, together with the boundary condition $\varphi(x) = \phi(x)$ on $\partial \Omega$, completely determines the macroscopic electric potential $\varphi(x)$. The following remarks are in order:

i. **Physical interpretation of the homogenized equation (9.26) for $\varphi(x)$**. Equation (9.26), together with the boundary condition $\varphi(x) = \phi(x)$ on $\partial \Omega$, corresponds to the governing equation for the electrostatic field within a homogeneous dielectric medium, with constant effective permittivity tensor $\hat{\varepsilon}$, which contains a distribution of space charges characterized by the effective space-charge density $\hat{q}(x)$.

ii. **The effective permittivity tensor $\hat{\varepsilon}$**. The effective permittivity tensor (9.27) that emerges in the homogenized equation (9.26) is independent of the choice of the domain $\Omega$ occupied by the composite, the boundary conditions on $\partial \Omega$, and the presence of space charges. Moreover, it follows from the properties (9.2) and (9.5) of the local permittivity $\varepsilon(y)$ and the definition (9.22) of the function $\omega_i(y)$ that $\hat{\varepsilon}$ satisfies the standard properties

$$\varepsilon_{ij} = \hat{\varepsilon}_{ji}, \quad \varepsilon_{ij} \xi_i \xi_j \geq \varepsilon_0 \xi_k \xi_k \ \forall \xi \in \mathbb{R}^N, \quad \hat{\varepsilon}_{ij} \in L^\infty(\Omega) \quad (9.29)$$

of a homogeneous dielectric medium; see, e.g., Section 2.3 of Chapter 1 in Bensoussan et al. (1978).

iii. **The effective space-charge density $\hat{q}(x)$**. The effective space-charge density (9.27) that emerges in the homogenized equation (9.26) is independent of the choice of the domain $\Omega$ occupied by
the composite and the boundary conditions on $\partial \Omega$. However, it does depend fundamentally on the presence of space charges through both of the constitutive functions $f(x)$ and $g(y)$ defining their density (9.10). It follows from the regularity (9.11) of the function $f(x)$ and the definiteness of the integrals in (9.28) that

$$\hat{q} \in H^1(\Omega).$$

(9.30)

It is also interesting to note that the total content of macroscopic space charges implied by the effective space-charge density (9.27)$_2$,

$$\int_{\Omega} \hat{q}(x) \, dx = - \int_{\Omega} \hat{\alpha}_{ij} \frac{\partial f_j}{\partial x_i}(x) \, dx,$$

(9.31)

need not be necessarily zero (only certain choices of the constitutive function $f(x)$ render macroscopic charge neutrality).

iv. Mathematical well-posedness. In view of the properties (9.29)$_{2,3}$ and (9.30) of $\hat{e}$ and $\hat{q}(x)$, and of the smoothness of $\partial \Omega$, it follows from the Lax-Milgram theorem that the solution of the homogenized equation (9.26), supplemented by the boundary condition $\varphi(x) = \phi(x)$ on $\partial \Omega$, for the macroscopic electric potential $\varphi(x)$ exists and is unique in $H^1(\Omega)$. The fact that the effective permittivity $\hat{e}$ is a constant together with the regularity $\phi \in H^{3/2}(\partial \Omega)$ and the smoothness of the boundary $\partial \Omega$ imply in fact the following stronger regularity result for $\varphi(x)$:

$$\varphi \in H^2(\Omega) \quad \text{and} \quad \text{Grad} \varphi \in H^1(\Omega; \mathbb{R}^N) \subset L^4(\Omega; \mathbb{R}^N);$$

(9.32)

see, e.g., Chapter 6.3 in Evans (2010) and Theorem 9.16 in Brezis (2011). The higher regularity $\varphi \in H^3(\Omega)$ can be obtained by considering boundary data $\phi \in H^{5/2}(\partial \Omega)$. We shall invoke this higher regularity in Section 9.3.

v. Computation of $\hat{e}$ and $\hat{q}(x)$. Evaluation of the formula (9.27)$_1$ for the effective permittivity tensor $\hat{e}$ requires knowledge of the $Y$-periodic function $\omega_i \in H^1_2(Y)$ defined by the pde (9.22)$_1$. In general, this pde does not admit an analytical solution and hence must be solved numerically; being linear elliptic, the pde (9.22)$_1$ can be readily solved, for instance, by the finite element method. Similarly, evaluation of the formula (9.27)$_2$ for the effective space-charge density $\hat{q}(x)$ requires knowledge of the $Y$-periodic function $\varpi_i \in H^1_2(Y)$ defined by the pde (9.22)$_2$. Given the alternative representation for $\hat{\alpha}_{ij}$ in the second equality of (9.28)$_2$ — which is a simple consequence of the divergence theorem and the $Y$-periodicity of the pdes (9.22) — the effective space-charge density $\hat{q}(x)$ can also be obtained directly from knowledge of $\omega_i(y)$. 


vi. The correction function $\varphi^{(1)}(x, y)$. Having completely determined the function $\varphi(x)$ in terms of equation (9.26) allows to determine (up to an additive function of $x$) the correction function $\varphi^{(1)}(x, y)$ in the expansion (9.13) by virtue of relation (9.23). Knowledge of $\varphi^{(1)}(x, y)$ allows in turn to determine the leading-order term of the corresponding asymptotic expansion for the electric field $E^\delta(X)$ in the limit as $\delta \to 0$:

$$E^\delta_i(X) = -\frac{\partial \varphi^\delta}{\partial X_i}(X) = \sum_{k=0}^{\infty} \delta^k E^{(k)}_i(x, y) = -\left(\frac{\partial \varphi}{\partial x_i}(x) + \frac{\partial \varphi^{(1)}}{\partial y_i}(x, y)\right) + O(\delta)$$

(9.33)

and, by the same token, the leading-order term of the expansion for the electric displacement field $D^\delta(X)$:

$$D^\delta_i(X) = \varepsilon_{ij} E^\delta_j(X) = \sum_{k=0}^{\infty} \delta^k D^{(k)}_j(x, y) = \varepsilon_{ij}(y) E^{(0)}_j(x, y) + O(\delta).$$

(9.34)

vii. The macro-variables. In addition to identifying $\varphi(x)$ as the macro-variable for the electric potential in the homogenized equation (9.26), a quick glance at (9.26) suffices to recognize the macroscopic electric field

$$E_i(x) \doteq -\frac{\partial \varphi}{\partial x_i}(x)$$

(9.35)

and the macroscopic electric displacement field

$$D_i(x) \doteq -\varepsilon_{ij} \frac{\partial \varphi}{\partial x_j}(x)$$

(9.36)

as the corresponding macro-variables that complete the electrostatics characterization of the resulting effective dielectric medium.

The macro-variable (9.35) turns out to be identical to the one that arises in the classical context of dielectric composites without rapidly oscillating source terms (see, e.g., Chapter 2 in Bensoussan et al., 1978). Namely, it corresponds to the average over the unit cell $Y$ of the leading-order term in the asymptotic expansion (9.33) of the electric field $E^\delta(X)$:

$$E_i(x) = \int_Y E^{(0)}_i(x, y) \, dy.$$

(9.37)

By the same token, the macro-variable (9.35) is consistent with the classical heuristic definition of macro-variables — in the absence of source terms — due to Hill (1963; 1972).
By contrast, the macro-variable (9.36) is not in accord with the classical result, instead relation (9.36) corresponds to the average over the unit cell $Y$ of the leading-order term in the asymptotic expansion (9.34) of the electric displacement field $\mathbf{D}^\delta(\mathbf{X})$ plus an additional contribution due to the presence of charges, specifically,

$$D_i(x) = \int_Y D_i^{(0)}(x,y) \, dy + \left( \int_Y \omega_i(y) g_j(y) \, dy \right) f_j(x). \quad (9.38)$$

viii. An alternative set of macro-variables. By exploiting the divergence form of the effective space-charge density (9.27) and rewriting the homogenized equation (9.26) as

$$\frac{\partial}{\partial x_i} \left[ -\bar{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(x) + \bar{\alpha}_{ij} f_j(x) \right] = 0, \quad (9.39)$$

one can alternatively define the same macroscopic electric field

$$E_i(x) = -\frac{\partial \varphi}{\partial x_i}(x) \quad (9.40)$$

as in remark vii above, but the different macro-variable

$$D_i(x) = -\bar{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(x) + \bar{\alpha}_{ij} f_j(x) \quad (9.41)$$

for the macroscopic electric displacement field instead of (9.36). Similar to the definition (9.36), the macro-variable (9.41) corresponds to the average over the unit cell $Y$ of the leading-order term in the asymptotic expansion (9.34) of the electric displacement field $\mathbf{D}^\delta(\mathbf{X})$ plus an additional contribution due to the presence of charges, in this case,

$$D_i(x) = \int_Y D_i^{(0)}(x,y) \, dy + \left( \int_Y y_i(y) g_j(y) \, dy \right) f_j(x). \quad (9.42)$$

Alternatively, this relation can be recast as a surface integral, namely,

$$D_i(x) = \int_{\partial Y} y_i D_j^{(0)}(x,y)n_j \, dy. \quad (9.43)$$

We conclude this remark by emphasizing that in the alternative view (9.39) of the homogenized equation (9.26), the homogenized material is no longer a standard homogenous dielectric that contains a distribution of space charges, but rather some sort of source-free polarized dielectric with the term $\bar{\alpha}_{ij} f_j(x)$ playing the role of an initial polarization in the electric displacement field.
9.2.2 The limit of the displacement field $u^\delta(X)$ as $\delta \to 0$

Next, we turn to the asymptotic analysis for the displacement field $u^\delta(X)$. Similar to the preceding asymptotic analysis for the electric potential field $\phi^\delta(X)$, it proves helpful to introduce the operators

$$B^\delta_{ik} = \delta^{-2}B^{(1)}_{ik} + \delta^{-1}B^{(2)}_{ik} + \delta^0B^{(3)}_{ik}$$

(9.44)

with

$$B^{(1)}_{ik} = \frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial}{\partial y_l} \right],$$

$$B^{(2)}_{ik} = \frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial}{\partial x_l} \right] + \frac{\partial}{\partial x_j} \left[ L_{ijkl}(y) \frac{\partial}{\partial y_l} \right],$$

$$B^{(3)}_{ik} = \frac{\partial}{\partial x_j} \left[ L_{ijkl}(y) \frac{\partial}{\partial x_l} \right],$$

(9.45)

and

$$C^\delta_i(h_1,h_2) = \delta^{-3}C^{(0)}_i(h_1,h_2) + \delta^{-2}C^{(1)}_i(h_1,h_2) + \delta^{-1}C^{(2)}_i(h_1,h_2) + \delta^0C^{(3)}_i(h_1,h_2)$$

(9.46)

with

$$C^{(0)}_i(h_1,h_2) = -\frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial y_l} \right],$$

$$C^{(1)}_i(h_1,h_2) = -\frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \left( \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial x_l} + \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial y_l} \right) \right] - \frac{\partial}{\partial x_j} \left[ M_{ijkl}(y) \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial y_l} \right],$$

$$C^{(2)}_i(h_1,h_2) = -\frac{\partial}{\partial x_j} \left[ M_{ijkl}(y) \left( \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial x_l} + \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial y_l} \right) \right] - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial x_l} \right],$$

$$C^{(3)}_i(h_1,h_2) = -\frac{\partial}{\partial x_j} \left[ M_{ijkl}(y) \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial x_l} \right]$$

(9.47)

in order to recast the pde (9.9) for the displacement field $u^\delta(X)$ in the more convenient form

$$B^\delta_{ik} u^\delta_k = C^\delta_i(\phi^\delta, \varphi^\delta).$$

(9.48)

Substituting the ansatz (9.13) in the pde (9.48) and expanding in powers of $\delta$ leads to a hierarchy of equations for the functions $u^{(k)}(x,y)$. Only the first four of these, of $O(\delta^{-3})$, $O(\delta^{-2})$, $O(\delta^{-1})$, and $O(\delta^0)$, turn out to be needed for our purposes here. In terms of the operators (9.45) and (9.47),
they read as

\[ 0 = C_i^0(\varphi^0, \varphi^0), \quad (9.49) \]

\[ B_{ik}^{(1)} u_k^{(0)} = C_i^1(\varphi^0, \varphi^0) + C_i^0(\varphi^0, \varphi^1) + C_i^0(\varphi^1, \varphi^0), \quad (9.50) \]

\[ B_{ik}^{(1)} u_k^{(1)} + B_{ik}^{(2)} u_k^{(0)} = C_i^2(\varphi^0, \varphi^0) + C_i^1(\varphi^0, \varphi^1) + C_i^1(\varphi^1, \varphi^0) + 
    C_i^0(\varphi^0, \varphi^2) + C_i^0(\varphi^2, \varphi^0) + C_i^0(\varphi^1, \varphi^1), \quad (9.51) \]

\[ B_{ik}^{(1)} u_k^{(2)} + B_{ik}^{(2)} u_k^{(1)} + B_{ik}^{(3)} u_k^{(0)} = C_i^3(\varphi^0, \varphi^0) + C_i^2(\varphi^0, \varphi^1) + C_i^2(\varphi^1, \varphi^0) + 
    C_i^1(\varphi^0, \varphi^2) + C_i^1(\varphi^2, \varphi^0) + C_i^1(\varphi^1, \varphi^1) + 
    C_i^0(\varphi^0, \varphi^3) + C_i^0(\varphi^3, \varphi^0) + C_i^0(\varphi^1, \varphi^2) + 
    C_i^0(\varphi^2, \varphi^1). \quad (9.52) \]

In connection with these equations, we emphasize that the function \( \varphi^0(x, y) = \varphi(x) \) has been completely determined in the preceding subsection, while the function \( \varphi^1(x, y) \) has been partially determined (up to an additive function of \( x \)). On the other hand, the functions \( \varphi^2(x, y) \) and \( \varphi^3(x, y) \) were not solved for since the relevant hierarchical equations were not considered. In the sequel, it will become evident that in spite of their appearance in equations (9.51) and (9.52), the functions \( \varphi^2(x, y) \) and \( \varphi^3(x, y) \) are actually not needed for our purposes here, namely, to work out the solution for the first two terms \( u^{(0)}(x, y) \) and \( u^{(1)}(x, y) \) in the expansion (9.13).2

The equation of order \( \delta^{-3} \)  

Granted the fact that the macroscopic electric potential \( \varphi^0(x, y) = \varphi(x) \) is independent of \( y \), the equation (9.49) of leading order is trivially satisfied.

The equation of order \( \delta^{-2} \)  

By invoking again the independence of \( \varphi(x) \) on \( y \), the equation (9.50) of order \( \delta^{-2} \) reduces to a pde for the function \( u^{(0)}(x, y) \) where \( y \) is the independent variable and \( x \) plays the role of a parameter. We write its unique solution (with respect to \( y \)) as

\[ u^{(0)}(x, y) = u(x). \quad (9.53) \]
The equation of order $\delta^{-1}$ Next, the equation (9.51) of order $\delta^{-1}$ can be written as

$$
\frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial u_{lp}(y)}{\partial y_l}(x, y) \right] = - \frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial u_k}{\partial x_l}(x) \right] - 
$$

$$
\frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \left( \frac{\partial \varphi^{(1)}(x, y)}{\partial y_k}(x) + \frac{\partial \varphi^{(1)}(x)}{\partial y_l}(x, y) \right) \right] - 
$$

$$
\frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \frac{\partial \varphi^{(1)}(x, y)}{\partial y_k}(x, y) \right] - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \frac{\partial \varphi^{(1)}(x, y)}{\partial y_l}(x, y) \right], \ y \in Y. \quad (9.54)
$$

For a given function $u(x)$ and a given $x$, this equation is a pde for the function $u^{(1)}(x, y)$ in the periodic unit cell $Y$ with $x$ playing the role of a parameter. With help of the representation (9.23) for the function $\varphi^{(1)}(x, y)$ and the introduction of the $Y$-periodic functions $\chi_{ijk}(y), \chi^{(1)}_{ijk}(y), \chi^{(2)}_{ijk}(y), \chi^{(3)}_{ijk}(y), \chi^{(4)}_{ijk}(y)$ defined implicitly as the unique solutions of the following linear elliptic pdes for $y \in Y$,

$$
\begin{align*}
\frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial \chi_{kpq}(y)}{\partial y_l}(y) \right] &= - \frac{\partial L_{ijpq}(y)}{\partial y_j}, \\
\int_Y \chi_{kpq}(y) dy &= 0 \\
\frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial \varphi^{(1)}(y)}{\partial y_l}(y) \right] &= - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \left( \delta_{kp} - \frac{\partial \omega_p(y)}{\partial y_k}(y) \right) \left( \delta_{lq} - \frac{\partial \omega_q(y)}{\partial y_l}(y) \right) \right], \\
\int_Y \chi^{(1)}_{kpq}(y) dy &= 0 \\
\frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial \varphi^{(2)}(y)}{\partial y_l}(y) \right] &= \frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \left( \delta_{kp} - \frac{\partial \omega_p(y)}{\partial y_k}(y) \right) \frac{\partial \omega_q(y)}{\partial y_l}(y) \right], \\
\int_Y \chi^{(2)}_{kpq}(y) dy &= 0 \\
\frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial \varphi^{(3)}(y)}{\partial y_l}(y) \right] &= \frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \frac{\partial \omega_p(y)}{\partial y_k}(y) \left( \delta_{lq} - \frac{\partial \omega_q(y)}{\partial y_l}(y) \right) \right], \\
\int_Y \chi^{(3)}_{kpq}(y) dy &= 0 \\
\frac{\partial}{\partial y_j} \left[ L_{ijkl}(y) \frac{\partial \varphi^{(4)}(y)}{\partial y_l}(y) \right] &= - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(y) \frac{\partial \omega_p(y)}{\partial y_k}(y) \frac{\partial \omega_q(y)}{\partial y_l}(y) \right], \\
\int_Y \chi^{(4)}_{kpq}(y) dy &= 0
\end{align*}
$$

(9.55)

the unique solution (with respect to $y$) of (9.54) can be written in the form

$$
\begin{align*}
u_{1}^{(1)}(x, y) &= \chi_{ipq}(y) \frac{\partial u_{ip}(x)}{\partial x_q}(x) + \chi^{(1)}_{ipq}(y) \frac{\partial \varphi}{\partial x_p}(x) + \chi^{(2)}_{ipq}(y) \frac{\partial \varphi}{\partial x_q}(x)f_q(x) + \\
&\chi^{(3)}_{ipq}(y)f_p(x) \frac{\partial \varphi}{\partial x_q}(x) + \chi^{(4)}_{ipq}(y)f_p(x)f_q(x) + s_{1}^{(1)}(x), \\
\end{align*}
$$

(9.56)

where $s_{1}^{(1)}(x)$ is an arbitrary function of $x$. 
The equation of order $\delta^0$ 

For a given function $u(x)$ and a given $x$ (since $u^{(1)}(x,y)$ is given by (9.56) in terms of $u(x)$), the equation (9.52) of order $\delta^0$ can be thought of as a pde for the function $u^{(2)}(x,y)$ in the periodic unit cell $Y$ with $x$ playing the role of a parameter. By invoking yet again the Fredholm alternative, such a pde admits a solution (with respect to $y$ and unique up to an additive constant) for $u^{(2)}(x,y)$ so long as the condition

$$
\int_Y \frac{\partial}{\partial x_j} \left[ L_{ijkl}(y) \left( \frac{\partial u_k}{\partial x_l}(x) + \frac{\partial u^{(1)}_k}{\partial y_l}(x,y) \right) \right] \, dy + \\
\int_Y \frac{\partial}{\partial x_j} \left[ M_{ijkl}(y) \left( \frac{\partial \varphi}{\partial x_k}(x) + \frac{\partial \varphi^{(1)}}{\partial y_k}(x,y) \right) \left( \frac{\partial \varphi}{\partial x_l}(x) + \frac{\partial \varphi^{(1)}}{\partial y_l}(x,y) \right) \right] \, dy = 0,
$$

(9.57)

is satisfied; in the derivation of this condition, use has been made of the divergence theorem together with the $Y$-periodicity of $L(y)$, $M(y)$, $\varphi^{(1)}(x,y)$, $\varphi^{(2)}(x,y)$, and $u^{(1)}(x,y)$.

Making now use of the representation (9.23) for $\varphi^{(1)}(x,y)$ in terms of the $Y$-periodic functions $\omega_i(y)$, $\varpi_i(y)$, the representation (9.56) for $u^{(1)}(x,y)$ in terms of the $Y$-periodic functions $\chi_{ijk}(y)$, $\bar{\chi}_{ijk}(y)$, $\bar{\chi}_{ijk}(y)$, $\bar{\chi}_{ijk}(y)$, and repeated use of the divergence theorem, equation (9.57) simplifies to

$$
\frac{\partial}{\partial x_j} \left[ \bar{L}_{ijkl}(x) \frac{\partial u_k}{\partial x_l}(x) + \bar{M}_{ijkl}(x) \frac{\partial \varphi}{\partial x_l}(x) \right] = -\bar{b}_i(x; \varphi(x)),
$$

(9.58)

where

$$
\bar{L}_{ijkl} = \int_Y L_{ijpq}(y) \left( \delta_{pk} \delta_{ql} + \frac{\partial \chi_{pkl}}{\partial y_q}(y) \right) \, dy,
$$

(9.59)

$$
\bar{M}_{ijkl} = \int_Y \left\{ L_{ijpq}(y) \frac{\partial \chi^{(1)}_{pkl}}{\partial y_q}(y) + M_{ijpq}(y) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(y) \right) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(y) \right) \right\} \, dy
$$

(9.60)

$$
= \int_Y M_{rsqp}(y) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rsi}}{\partial y_s}(y) \right) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(y) \right) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(y) \right) \, dy,
$$

and

$$
\bar{b}_i(x; \varphi(x)) = \frac{\partial}{\partial x_j} \left[ -\bar{B}_{ijkl}(x) f_i(x) - \bar{B}_{ijkl}^{(2)} f_k(x) \frac{\partial \varphi}{\partial x_l}(x) + \bar{B}_{ijkl}^{(3)} f_k(x) f_i(x) \right]
$$

(9.61)

with
\[
\hat{B}_{ijkl}^{(1)} = -\int_Y \left\{ L_{ijpq}(y) \frac{\partial \chi_{pkl}}{\partial y_q}(y) - M_{ijpq}(y) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(y) \right) \frac{\partial \varpi_l}{\partial y_q}(y) \right\} \, dy \\
= \int_Y M_{rspq}(y) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(y) \frac{\partial \varpi_k}{\partial y_p}(y) \right) \left( \delta_{ql} - \frac{\partial \varpi_l}{\partial y_q}(y) \right) \, dy,
\]

\[
\hat{B}_{ijkl}^{(2)} = -\int_Y \left\{ L_{ijpq}(y) \frac{\partial \chi_{pkl}}{\partial y_q}(y) - M_{ijpq}(y) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(y) \right) \frac{\partial \varpi_l}{\partial y_q}(y) \right\} \, dy \\
= \int_Y M_{rspq}(y) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(y) \frac{\partial \varpi_k}{\partial y_p}(y) \right) \left( \delta_{ql} - \frac{\partial \varpi_l}{\partial y_q}(y) \right) \, dy,
\]

\[
\hat{B}_{ijkl}^{(3)} = \int_Y \left\{ L_{ijpq}(y) \frac{\partial \chi_{pkl}}{\partial y_q}(y) + M_{ijpq}(y) \frac{\partial \varpi_k}{\partial y_p}(y) \frac{\partial \varpi_i}{\partial y_q}(y) \right\} \, dy \\
= \int_Y M_{rspq}(y) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(y) \frac{\partial \varpi_k}{\partial y_p}(y) \right) \left( \delta_{ql} - \frac{\partial \varpi_l}{\partial y_q}(y) \right) \, dy.
\]

Figure 9.2: Schematic illustrating that in the limit as \( \delta \to 0 \) an elastic dielectric composite with heterogeneous properties \( \varepsilon^\delta(X), L^\delta(X), M^\delta(X) \) containing a distribution of passive space charges — characterized by the space-charge density (9.10) with arbitrary but fixed functions \( f(X) \) and \( g(\delta^{-1}X) \) — reduces to an elastic dielectric with homogeneous effective properties \( \hat{\varepsilon}, \hat{L}, \hat{M} \), effective space-charge density \( \hat{q}(X) \), and effective body-force density \( \hat{b}(X; \varphi(X)) \).

For a given macroscopic electric field \( \varphi(x) \) defined by the pde (9.26) and a given boundary condition \( u(x) = v(x) \) on \( \partial \Omega \), equation (9.58) is the homogenized pde in \( \Omega \) that completely determines the macroscopic displacement field \( u(x) \). The following remarks are in order:

i. **Physical interpretation of the homogenized equation (9.58) for \( u(x) \).** Equation (9.58), together with the one-way coupled pde (9.26) for the macroscopic electric field \( \varphi(x) \) and the boundary conditions \( \varphi(x) = \phi(x) \) and \( u(x) = v(x) \) on \( \partial \Omega \), constitutes the governing equation for the displacement field within a homogeneous elastic dielectric medium, with constant effective elasticity tensor \( \hat{L} \) and constant effective electrostriction tensor \( \hat{M} \), which contains a distribution...
of body forces characterized by the effective body-force density \( \hat{b}(x; \varphi(x)) \). Figure 9.2 provides a schematic of this physical interpretation of the homogenized equations (9.26) and (9.58).

ii. The effective elasticity and electrostriction tensors \( \tilde{L} \) and \( \hat{M} \). Much like the effective permittivity tensor \( \tilde{\varepsilon} \) in the homogenized equation (9.26) for \( \varphi(x) \), the effective elasticity tensor (9.59) and the effective electrostriction tensor (9.60) in the homogenized equation (9.58) for \( u(x) \) are independent of the choice of the domain \( \Omega \) occupied by the composite, the boundary conditions on \( \partial \Omega \), and the presence of space charges. Moreover, it follows from the properties (9.3)_2 and (9.5)_2 of the local elasticity tensor \( L(y) \) and the definition (9.55)_1 of the function \( \chi_{ijk}(y) \) that \( \tilde{L} \) satisfies the standard properties

\[
\tilde{L}_{ijkl} = \tilde{L}_{klij} = \tilde{L}_{ijlk} > \theta \Xi_{pq} \Xi_{pq} \quad \forall \Xi \in \mathbb{R}^{N \times N}, \quad \tilde{L}_{ijkl} \in L^\infty(\Omega),
\]

for some positive constant \( \theta \), of a homogeneous elastic dielectric medium; see, e.g., Section 2.3 of Chapter 1 in Bensoussan et al. (1978) and Chapter 1.1.4 in Allaire (2002). Similarly, it follows from the properties (9.3)_2 and (9.5)_2 of the local elasticity tensor \( L(y) \), the property (9.5)_3 of the local electrostriction tensor \( M(y) \), and the definition (9.55)_1 of the function \( \chi_{ijk}(y) \) that \( \hat{M} \) also satisfies the standard properties

\[
\hat{M}_{ijkl} = \hat{M}_{ijlk} = \hat{M}_{ijlk} \in L^\infty(\Omega)
\]

of a homogeneous elastic dielectric medium.

iii. The effective body-force density \( \hat{b}(x; \varphi(x)) \). In spite of the fact that there are no body forces in the original boundary-value problem (9.9) for \( u^\delta(X) \), body forces appear in the homogenized equation (9.58) for \( u(x) \). These emerge as a result of the presence of space charges in the coupled boundary-value problem (9.8) for \( \varphi^\delta(X) \). In particular, the effective body-force density (9.61) that emerges in the homogenized equation (9.58) is independent of the choice of the domain \( \Omega \) occupied by the composite and the boundary conditions on \( \partial \Omega \). Its dependence on the presence of space charges is through both constitutive functions \( f(x) \) and \( g(y) \) defining their density (9.10), as well as through the macroscopic electric potential \( \varphi(x) \). Granted the boundedness (9.5)_3 of the local electrostriction tensor \( M(y) \), the regularity (9.11)_1 of the constitutive function \( f(x) \), the definitions (9.22) and (9.55)_1 of the functions \( \omega_i(y) \), \( \varpi_i(y) \), \( \chi_{ijk}(y) \), and the fact that \( \text{Grad} \varphi \in H^1(\Omega; \mathbb{R}^N) \subset L^4(\Omega; \mathbb{R}^N) \), it follows that the effective
body-force density (9.61) is of divergence form, in particular,

\[ \hat{b}(x; \varphi(x)) = \text{Div} \mathbf{B}(x), \quad \mathbf{B} \in L^2(\Omega; \mathbb{R}^{N \times N}). \]  

(9.67)

iv. **Mathematical well-posedness.** Granted the properties (9.65)\_2,3, (9.66)\_2, (9.67) of \( \hat{L}, \hat{M}, \hat{b}(x; \varphi(x)) \), the regularity result (9.32)\_2 for \( \varphi(x) \), the boundary condition \( u(x) = v(x) \in H^{1/2}(\partial \Omega; \mathbb{R}^N) \) on \( \partial \Omega \), and the smoothness of \( \partial \Omega \), it follows from the Lax-Milgram theorem that the solution of the homogenized equation (9.58) for the macroscopic displacement field \( u(x) \) exists, is unique, and

\[ u \in H^1(\Omega; \mathbb{R}^N). \]  

(9.68)

v. **Computation of \( \hat{L}, \hat{M}, \) and \( \hat{b}(x; \varphi(x)) \).** Evaluation of the formulas (9.59) and (9.60) for the effective elasticity tensor \( \hat{L} \) and the effective electrostriction tensor \( \hat{M} \) requires knowledge of the \( Y \)-periodic function \( \chi_{ipq} \in H^1_\#(Y) \) defined by the pde (9.55)\_1 and the \( Y \)-periodic function \( \omega_i \in H^1_\#(Y) \) defined by the pde (9.22)\_1. These are linear elliptic pdes that do not generally admit an analytical solution, but they can be readily solved numerically, for instance, by the finite element method. In addition to the solution of the homogenized equation (9.26) for the macroscopic electric potential \( \varphi(x) \), evaluation of the formula (9.61) for the effective body-force density \( \hat{b}(x; \varphi(x)) \) also requires the solutions for the functions \( \omega_i \in H^1_\#(Y) \) and \( \chi_{ijk} \in H^1_\#(Y) \), as well as for the \( Y \)-periodic function \( \varpi_i \in H^1_\#(Y) \) defined by the linear elliptic pde (9.22)\_2, whose gradients appear in the tensors \( \hat{B}^{(1)}, \hat{B}^{(2)}, \text{and } \hat{B}^{(3)}. \)

vi. **The correction function \( u^{(1)}(x, y) \).** By virtue of relation (9.56), having completely determined the function \( \varphi(x) \) from equation (9.26) and the function \( u(x) \) from equation (9.58) allows to determine (up to an additive function of \( x \)) the correction function \( u^{(1)}(x, y) \) in the expansion (9.13)\_2. Knowledge of \( u^{(1)}(x, y) \) allows in turn to determine the leading-order term in the corresponding expansion for the gradient of the displacement field \( \mathbf{H}^\delta(X) \) in the limit as \( \delta \rightarrow 0: \)

\[ H^\delta_{ij}(X) = \frac{\partial u^\delta_i}{\partial X_j}(X) = \sum_{k=0}^\infty \delta^k H^{(k)}_{ij}(x, y) = \frac{\partial u_i}{\partial x_j}(x) + \frac{\partial u^{(1)}_i}{\partial y_j}(x, y) + O(\delta) \]  

(9.69)

and, by the same token, the leading-order term of the expansion for the first Piola-Kirchhoff
stress tensor $S^δ(\mathbf{X})$:

$$S^δ_{ij}(\mathbf{X}) = L^δ_{ijkl}(\mathbf{X})H^δ_{kl}(\mathbf{X}) + M^δ_{ijkl}(\mathbf{X})E^δ_k(\mathbf{X})E^δ_l(\mathbf{X}) = \sum_{k=0}^{\infty} \delta^k S^{(k)}_{ij}(\mathbf{x}, \mathbf{y}) = L_{ijkl}(\mathbf{y})H^{(0)}_{kl}(\mathbf{x}, \mathbf{y}) + M_{ijkl}(\mathbf{y})E^{(0)}_k(\mathbf{x}, \mathbf{y}) E^{(0)}_l(\mathbf{x}, \mathbf{y}) + O(\delta) \quad (9.70)$$

vii. The macro-variables. Similar to the identification of macro-variables in the homogenized equation (9.26) for the macroscopic electric potential $\varphi(\mathbf{x})$, a quick glance at (9.58) suffices to recognize not only $\mathbf{u}(\mathbf{x})$ as the macro-variable for the displacement field, but also the macroscopic gradient of the displacement

$$H_{ij}(\mathbf{x}) = \frac{\partial u_i}{\partial x_j}(\mathbf{x}) \quad (9.71)$$

and the macroscopic stress

$$S_{ij}(\mathbf{x}) = \bar{L}_{ijkl} \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \bar{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) \quad (9.72)$$

as the corresponding macro-variables that complete the electroelastostatics characterization of the resulting effective elastic dielectric medium.

Akin to the standard macro-variable (9.35) that arises for the electric field, (9.71) is the standard macro-variable that emerges in the classical context of linear elasticity without rapidly oscillating source terms (see, e.g., Chapter 1 in Allaire, 2002), in the sense that it corresponds to the average over the unit cell $Y$ of the leading-order term in the asymptotic expansion (9.69) of the gradient of the displacement field $H^δ(\mathbf{X})$:

$$H_{ij}(\mathbf{x}) = \int_Y H^{(0)}_{ij}(\mathbf{x}, \mathbf{y}) d\mathbf{y}. \quad (9.73)$$

The macro-variable (9.71) is then also in accord with the classical heuristic definition of macro-variables (Hill, 1963; Hill, 1972).

By contrast, the macro-variable (9.72) differs from the classical result, since it corresponds to the average over the unit cell $Y$ of the leading-order term in the asymptotic expansion (9.70) of the stress $S^δ(\mathbf{X})$ plus additional contributions due to the presence of charges, specifically,

$$S_{ij}(\mathbf{x}) = \int_Y S^{(0)}_{ij}(\mathbf{x}, \mathbf{y}) d\mathbf{y} + \hat{B}^{(1)}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) f_l(\mathbf{x}) + \hat{B}^{(2)}_{ijkl} f_k(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) - \hat{B}^{(3)}_{ijkl} f_k(\mathbf{x}) f_l(\mathbf{x}). \quad (9.74)$$
viii. An alternative set of macro-variables. By exploiting the divergence form of the effective body-force density (9.61) and rewriting the homogenized equation (9.58) as

\[
\frac{\partial}{\partial x_j} \left[ L_{ijkl} \frac{\partial u_k}{\partial x_i} (x) + M_{ijkl} \frac{\partial \varphi}{\partial x_i} (x) \frac{\partial \varphi}{\partial x_k} (x) - B_{ijkl}^{(1)} \frac{\partial \varphi}{\partial x_k} (x) f_l (x) - B_{ijkl}^{(2)} f_k (x) \frac{\partial \varphi}{\partial x_l} (x) f_l (x) + B_{ijkl}^{(3)} f_k (x) f_l (x) \right] = 0,
\]

(9.75)

one can alternatively define the same macroscopic electric field

\[
H_{ij} (x) = \frac{\partial u_i}{\partial x_j} (x)
\]

(9.76)
as in remark vii above, but the different macro-variable

\[
S_{ij} (x) \equiv L_{ijkl} \frac{\partial u_k}{\partial x_i} (x) + M_{ijkl} \frac{\partial \varphi}{\partial x_i} (x) \frac{\partial \varphi}{\partial x_k} (x) - B_{ijkl}^{(1)} \frac{\partial \varphi}{\partial x_k} (x) f_l (x) - B_{ijkl}^{(2)} f_k (x) \frac{\partial \varphi}{\partial x_l} (x) f_l (x) + B_{ijkl}^{(3)} f_k (x) f_l (x)
\]

(9.77)

for the macroscopic stress instead of (9.72). Contrary to the definition (9.72), the macro-variable (9.77) is consistent with the standard definition that emerges in the classical context of linear elasticity without rapidly oscillating source terms (see, e.g., Chapter 1 in Allaire, 2002), in the sense that it corresponds to the average over the unit cell \( Y \) of the leading-order term in the asymptotic expansion (9.70) of the stress \( S^\delta (X) \):

\[
S_{ij} (x) = \int_Y S_{ij}^{(0)} (x, y) dy.
\]

(9.78)

In regard to the above-identified alternative set of macro-variables, we emphasize that in the alternative view (9.75) of the homogenized equation (9.58), the homogenized material is no longer a standard homogenous elastic dielectric with even electromechanical coupling that contains a distribution of space charges and body forces, but rather a source-free elastic dielectric with complicated electromechanical coupling which is neither even nor odd.

ix. The absence of space charges. In the absence of space charges when \( f(x) = 0 \) and/or \( g(y) = 0 \), the effective space-charge density (9.27)\(_2\) and the effective body-force density (9.61) vanish,

\[
\tilde{q}(x) = 0 \quad \text{and} \quad \tilde{b}(x; \varphi(x)) = 0,
\]

(9.79)

and the homogenized equations (9.26) and (9.58), supplemented by the boundary conditions \( \varphi(x) = \phi(x) \) and \( u(x) = v(x) \) on \( \partial \Omega \), reduce to the homogenized equations originally obtained by Tian (2007) and Tian et al. (2012) via the two-scale convergence method (Allaire, 1992); see expressions (2.33)–(2.35) with (2.36)–(2.37) in Section 2.2.1.
9.3 A class of active charges

The preceding derivation of the homogenized equations (9.26) and (9.58) for the macroscopic electric field \( \varphi(x) \) and macroscopic displacement field \( u(x) \) is valid for any choice — subject to the conditions (9.11) — of constitutive functions \( f(x) \) and \( g(y) \) defining the density of space charges (9.10) in the composite. These functions may be chosen not to be fixed or passive but to be active instead, in the sense that they may be selected to depend on \( \varphi^\delta(X) \) and/or on \( u^\delta(X) \). More generally, the functions \( f(x) \) and \( g(y) \) may be selected to have both passive as well as active components.

In this section, motivated by the work of Lopez-Pamies et al. (2014), we work out the homogenized equations for elastic dielectric composites that contain a special class of active charges wherein the function \( g(y) \) is arbitrary but fixed while the function \( f(x) \) is set to be proportional to the macroscopic electric field:

\[
    f_i(x) = -\frac{\partial \varphi}{\partial x_i}(x). \tag{9.80}
\]

From a mathematical point of view, we remark that this choice is valid provided that \( \varphi \in H^3(\Omega) \) since the function \( f(x) \) was chosen from the outset to have the regularity (9.11). Accordingly, throughout this section, we shall assume that the boundary data \( \phi \in H^{5/2}(\partial\Omega) \). As it will become clear further below, this will ensure that \( \varphi \in H^3(\Omega) \); cf. remark iv following the homogenized equation (9.26).

From a physical standpoint, roughly speaking, relation (9.80) corresponds to a microscopic distribution of space charges that scales in magnitude and aligns in direction with the electric field at the macroscopic material point \( x \). The precise details of the local alignment of the space charges are characterized by the specifics of the function \( g(y) \). At this point, it is important to emphasize that little is actually known about the constitutive behavior of active space charges in deformable solids from direct experimental measurements. Indeed, for the prominent case of dielectric elastomers filled with (semi-)conducting or high-dielectric nanoparticles (see, e.g., Huang et al., 2005; Meddeb and Ounaies, 2012; Liu et al., 2013), space charges are expected to be active (i.e., locally mobile) in the regions of the elastomer immediately surrounding the nanoparticles (Lewis, 2004; Roy et al., 2005; Nelson, 2010), but direct measurements of the precise content and local mobility of the space charges contained therein have proven thus far difficult. The prescription (9.80) corresponds perhaps to the simplest physically plausible prototype that is consistent with the otherwise accessible macroscopic experimental measurements (Lopez-Pamies et al., 2014; see also Chapter 8). In this regard, it is
also important to remark that other classes of active space charges — such as, for instance, those described by the local version \( f(\mathbf{X}) = \text{Grad} \varphi^a(\mathbf{X}) \) of (9.80) — have been checked to lead to similar results to those that ensue from (9.80), and hence support the general physical implications presented here.

Granted the constitutive choice (9.80) for \( f(\mathbf{x}) \), it is straightforward to deduce from (9.26) that the homogenized equation for the macroscopic electric field \( \varphi(\mathbf{x}) \) is given by

\[
\frac{\partial}{\partial x_i} \left( \tilde{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) \right) = 0 \quad (9.81)
\]

with

\[
\tilde{\varepsilon}_{ij} = \varepsilon_{ij} + \tilde{\alpha}_{ij} = \int_Y \left\{ \varepsilon_{ik}(y) \left( \delta_{jk} - \frac{\partial \omega_j(y)}{\partial y_k} + \frac{\partial \varpi_j(y)}{\partial y_k} \right) + y_i g_j(y) \right\} d\mathbf{y}. \quad (9.82)
\]

Similarly, it is straightforward to deduce from (9.58) that the homogenized equation for the macroscopic displacement field \( \mathbf{u}(\mathbf{x}) \) is given by

\[
\frac{\partial}{\partial x_j} \left[ \tilde{L}_{ijkl} \frac{\partial u_k}{\partial x_i}(\mathbf{x}) + \tilde{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) \right] = 0, \quad (9.83)
\]

where the macroscopic electric field \( \varphi(\mathbf{x}) \) is defined implicitly by (9.81), it is recalled that \( \tilde{L}_{ijkl} \) is given by expression (9.59),

\[
\tilde{M}_{ijkl} = \tilde{M}_{ijkl} + \tilde{B}^{(1)}_{ijkl} + \tilde{B}^{(2)}_{ijkl} + \tilde{B}^{(3)}_{ijkl} = \int_Y \tilde{M}_{rsrp}(\mathbf{y}) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s(\mathbf{y})} \right) \times
\]

\[
\left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(\mathbf{y}) + \frac{\partial \varpi_k}{\partial y_p}(\mathbf{y}) \right) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(\mathbf{y}) + \frac{\partial \varpi_l}{\partial y_q}(\mathbf{y}) \right) d\mathbf{y}, \quad (9.84)
\]

and it is further recalled that \( \omega_i(\mathbf{y}), \varpi_i(\mathbf{y}), \) and \( \chi_{ijk}(\mathbf{y}) \) are the \( Y \)-periodic functions with zero average in \( Y \) defined by the pdes (9.22) and (9.55).

Equations (9.81) and (9.83), together with the boundary conditions \( \varphi(\mathbf{x}) = \phi(\mathbf{x}) \) and \( \mathbf{u}(\mathbf{x}) = \mathbf{v}(\mathbf{x}) \) on \( \partial \Omega \), are the homogenized pdes in \( \Omega \) for the macroscopic electric field \( \varphi(\mathbf{x}) \) and macroscopic displacement field \( \mathbf{u}(\mathbf{x}) \). A number of remarks are in order:

i. **Physical interpretation of the homogenized equations (9.81) and (9.83) for \( \varphi(\mathbf{x}) \) and \( \mathbf{u}(\mathbf{x}) \).** The one-way coupled system of pdes (9.81) and (9.83) for the macroscopic electric field \( \varphi(\mathbf{x}) \) and the macroscopic displacement field \( \mathbf{u}(\mathbf{x}) \) constitute the governing equations for a homogeneous elastic dielectric medium, with constant effective permittivity tensor \( \tilde{\varepsilon} \), constant effective elasticity tensor \( \tilde{\mathbf{L}} \), and constant effective electrostriction tensor \( \tilde{\mathbf{M}} \). Remarkably, in spite of the fact that the elastic dielectric composite contains a distribution of space charges at the length
9. Homogenization of elastic dielectric composites with rapidly oscillating source terms

Figure 9.3: Schematic illustrating that in the limit as $\delta \to 0$ an elastic dielectric composite with heterogeneous properties $\epsilon(\mathbf{X}), L(\mathbf{X}), M(\mathbf{X})$ containing a distribution of active space charges — characterized by the density (9.10) with the choice (9.80) for the function $\mathbf{f}(\mathbf{X})$ and arbitrary but fixed function $\mathbf{g}(\delta^{-1}\mathbf{X})$ — reduces to an elastic dielectric with homogeneous effective properties $\tilde{\epsilon}, \tilde{L}, \tilde{M}$.

scale of the microstructure, neither an effective space-charge density nor an effective body-force density show up in the homogenized equations (9.81) and (9.83). Instead, the effect of the space charges shows up in the effective permittivity tensor $\tilde{\epsilon}$ and the effective electrostriction tensor $\tilde{M}$; this distinctive feature, which is in direct contrast to the result obtained for passive charges in the preceding section, is elaborated further in the next remarks. Figure 9.3 provides a schematic of the above-identified physical interpretation of the homogenized equations (9.81) and (9.83).

ii. The effective permittivity, elasticity, and electrostriction tensors $\tilde{\epsilon}, \tilde{L}, \tilde{M}$. The effective elasticity tensor $\tilde{L}$ in the homogenized equation (9.83) is identical to the effective elasticity tensor in the homogenized equation (9.58) for the case of passive charges; its properties are outlined in remark ii of Section 9.2.2. On the other hand, the effective permittivity tensor (9.82) and the effective electrostriction tensor (9.84) that emerge in the homogenized equations (9.81) and (9.83) are different from their counterparts in (9.26) and (9.58). While they are independent of the choice of the domain $\Omega$ occupied by the composite and the boundary conditions on $\partial \Omega$, the effective tensors (9.82) and (9.84) do depend strongly on the presence of space charges through the constitutive function $\mathbf{g}(\mathbf{y})$, which, as discussed in Section 9.1, describes the distribution of space charges at the length scale of the microstructure.

More specifically, it follows from the regularity (9.5) of the local permittivity tensor $\epsilon(\mathbf{y})$, the
definitions \((9.22)\) of the functions \(\omega_i(y)\) and \(\varpi_i(y)\), and the boundedness \((9.11)_3\) of \(g(y)\) that the effective permittivity \((9.82)\) is bounded,

\[
\tilde{\varepsilon}_{ij} \in L^\infty(\Omega),
\]

but, rather remarkably, it is \emph{not} necessarily symmetric, \emph{nor} positive definite for the cases when is symmetric; whether \(\tilde{\varepsilon}\) is symmetric and, if so, positive definite, depends on the choice of constitutive function \(g(y)\). It further follows from the properties \((9.4)\) and \((9.5)_3\) of the local electrostriction tensor \(M(y)\), together with the definitions \((9.22)\) and \((9.55)_1\) of the functions \(\omega_i(y)\), \(\varpi_i(y)\), and \(\chi_{ijk}(y)\) that the effective electrostriction tensor \((9.84)\) satisfies the standard properties

\[
\tilde{M}_{ijkl} = \tilde{M}_{jikl} = \tilde{M}_{ijlk}, \quad \tilde{M}_{ijkl} \in L^\infty(\Omega)
\]

of a homogeneous elastic dielectric medium.

Here, it is important to recognize that in spite of the boundedness \((9.85)\) and \((9.86)_2\), the components of the effective permittivity \((9.82)\) and the effective electrostriction tensor \((9.84)\) can be made to achieve arbitrarily large positive or negative values as, in essence, they are proportional to the constitutive function \(g(y)\). The physical implications of these features are far reaching. Indeed, these features confirm that judicious manipulation of space charges in deformable dielectric composites provides a promising path forward for the design of materials with exceptional electromechanical properties, including materials with unusually large permittivities and electrostriction coefficients and metamaterials featuring negative permittivity (see, e.g., Smith et al., 2015).

\textit{iii. Mathematical well-posedness.} For choices of the constitutive function \(g(y)\) that lead to effective permittivity tensors \(\tilde{\varepsilon}\) that are symmetric and either positive or negative definite, the homogenized equation \((9.81)\) for the macroscopic electric field \(\varphi(x)\) is nothing more than a standard second-order linear elliptic pde with constant coefficient and hence, given the boundary condition \(\varphi(x) = \phi(x) \in H^{5/2}(\partial \Omega)\) on \(\partial \Omega\) and the smoothness of \(\partial \Omega\), its solution exists, is unique, and possesses the following regularity properties:

\[
\varphi \in H^3(\Omega) \quad \text{and} \quad \text{Grad} \varphi \in H^2(\Omega; \mathbb{R}^N) \subset L^4(\Omega; \mathbb{R}^N).
\]  

\((9.87)\)

In turn, granted the properties \((9.65)_{2,3}\) and \((9.86)_2\) of \(\tilde{L}\) and \(\tilde{M}\), the regularity result \((9.87)_2\) for \(\varphi(x)\), the boundary condition \(u(x) = v(x) \in H^{1/2}(\partial \Omega; \mathbb{R}^N)\) on \(\partial \Omega\), and the smoothness of \(\partial \Omega\),
the Lax-Milgram theorem ensures existence and uniqueness of the solution of the homogenized equation (9.83) for the macroscopic displacement field \( u(x) \), in particular,

\[
u \in H^1(\Omega; \mathbb{R}^N).
\] (9.88)

For choices of the constitutive function \( g(y) \) that lead to effective permittivity tensors \( \tilde{\varepsilon} \) that are not symmetric but satisfy the ellipticity condition \( \varepsilon_{ij} \xi_i \xi_j \geq \varepsilon_0 \xi_k \xi_k \forall \xi \in \mathbb{R}^N \), solutions for the macroscopic fields \( \varphi(x) \) and \( u(x) \) also exist, are unique, and possess the regularity (9.87) and (9.88). Finally, for choices of the constitutive function \( g(y) \) that lead to effective permittivity tensors \( \tilde{\varepsilon} \) that (are either symmetric or not symmetric but) do not satisfy the ellipticity condition \( \varepsilon_{ij} \xi_i \xi_j \geq \varepsilon_0 \xi_k \xi_k \forall \xi \in \mathbb{R}^N \), the homogenized equation (9.81) is not elliptic and hence solutions for the macroscopic electric field \( \varphi(x) \) may not exist.

iv. Computation of \( \tilde{\varepsilon} \), \( \tilde{\mathbf{L}} \), and \( \tilde{\mathbf{M}} \). Evaluation of the formulas (9.82) and (9.84) for the effective permittivity tensor \( \tilde{\varepsilon} \) and effective electrostriction tensor \( \tilde{\mathbf{M}} \) requires knowledge of the \( Y \)-periodic functions \( \omega_i \in H^1_\#(Y) \), \( \varpi_i \in H^1_\#(Y) \) only through the linear combination \( \varpi_i(y) - \omega_i(y) \). Their evaluation amounts then to solving not the two boundary-value problems (9.22) for \( \omega_i(y) \) and \( \varpi_i(y) \), but instead the single boundary-value problem

\[
\begin{cases}
\frac{\partial}{\partial y_i} \left[ \varepsilon_{ik}(y) \left( \delta_{kj} + \frac{\partial \varpi_j}{\partial y_k}(y) \right) \right] = g_j(y), \quad y \in Y \\
\int_Y \dot{\omega}_j(y) dy = 0
\end{cases}
\]

for \( \dot{\omega}_i(y) = \varpi_i(y) - \omega_i(y) \). (9.89)

Evaluation of the formula (9.59) for the effective elasticity tensor \( \tilde{\mathbf{L}} \) requires knowledge of the \( Y \)-periodic function \( \chi_{ijk} \in H^1_\#(Y) \) defined by the pde (9.55)_1. The two above-identified equations for \( \dot{\omega}_i(y) \) and \( \chi_{ijk}(y) \) are linear elliptic pdes that can be readily solved numerically, for instance, again, by the finite element method. An example of such a finite-element numerical solution is presented further below in Section 9.4.

v. The leading-order terms for the electric field \( \mathbf{E}^\delta(X) \), the electric displacement field \( \mathbf{D}^\delta(X) \), the gradient of the displacement field \( \mathbf{H}^\delta(X) \), and the stress \( \mathbf{S}^\delta(X) \). Granted the constitutive choice (9.80) for the function \( f(x) \), it is a simple matter to deduce from (9.33) and (9.34) that the leading-order terms in the homogenization limit as \( \delta \to 0 \) for the electric field \( \mathbf{E}^\delta(X) \) and the electric displacement field \( \mathbf{D}^\delta(X) \) specialize to

\[
E_i^{(0)}(x,y) = - \left( \delta_{ik} - \frac{\partial \omega_k}{\partial y_i}(y) + \frac{\partial \varpi_k}{\partial y_i}(y) \right) \frac{\partial \varphi}{\partial x_k}(x)
\] (9.90)
and
\[ D_i^{(0)}(x, y) = \varepsilon_{ij}(y) E_j^{(0)}(x, y). \] (9.91)

Similarly, with help of the notation \( \tilde{\chi}_{kpq}(y) = \chi_{kpq}^{(1)}(y) - \chi_{kpq}^{(2)}(y) - \chi_{kpq}^{(3)}(y) + \chi_{kpq}^{(4)}(y) \), it is a simple matter to deduce from (9.69) and (9.70) that the leading-order terms for the gradient of the displacement field \( H^{(0)}(x) \) and the first Piola-Kirchhoff stress tensor \( S^{(0)}(x) \) specialize to
\[ H_{ij}^{(0)}(x, y) = \frac{\partial u_i}{\partial x_j}(x) + \frac{\partial \tilde{\chi}_{ipq}}{\partial y_j}(y) \frac{\partial u_p}{\partial x_q}(x) + \frac{\partial \tilde{\chi}_{ipq}}{\partial y_q}(y) \frac{\partial \phi}{\partial x_p}(x), \] (9.92)
and
\[ S_{ij}^{(0)}(x, y) = L_{ijkl}(y) H_{kl}^{(0)}(x, y) + M_{ijkl}(y) E_k^{(0)}(x, y) E_l^{(0)}(x, y). \] (9.93)

vi. The macro-variables. The macro-variables that emerge from the one-way coupled homogenized equations (9.81) and (9.83) for the macroscopic electric field and the macroscopic electric displacement field can be readily deduced to be given by
\[ E_i(x) = -\frac{\partial \phi}{\partial x_i}(x) \quad \text{and} \quad D_i(x) = -\tilde{\varepsilon}_{ij} \frac{\partial \phi}{\partial x_j}(x), \] (9.94)
while those that emerge for the macroscopic gradient of the displacement field and the macroscopic stress are given by
\[ H_{ij}(x) = \frac{\partial u_i}{\partial x_j}(x) \quad \text{and} \quad S_{ij}(x) = \tilde{L}_{ijkl}(x) \frac{\partial u_k}{\partial x_l}(x) + \tilde{M}_{ijkl}(x) \frac{\partial \phi}{\partial x_k}(x) \frac{\partial \phi}{\partial x_l}(x). \] (9.95)

The macro-variables (9.94) for the electric field, the gradient of the deformation, and the stress agree with the classical definition,
\[ E_i(x) = \int_Y E_i^{(0)}(x, y) \, dy, \quad H_{ij}(x) = \int_Y H_{ij}^{(0)}(x, y) \, dy, \quad S_{ij}(x) = \int_Y S_{ij}^{(0)}(x, y) \, dy, \] (9.96)
while the macro-variable (9.94) for the electric displacement field does not. Instead, relation (9.94) corresponds to the average over the unit cell \( Y \) of the leading-order term in the asymptotic expansion (9.91) of the electric displacement field \( D^{(0)}(X) \) plus an additional contribution due to the presence of charges, specifically,
\[ D_i(x) = \int_Y D_i^{(0)}(x, y) \, dy - \left( \int_Y y_i g_j(y) \, dy \right) \frac{\partial \phi}{\partial x_j}(x). \] (9.97)
As opposed to its counterpart (9.38) for the case of passive charges, relation (9.97) can be written as a surface integral:
\[ D_i(x) = \int_{\partial Y} y_i D_i^{(0)}(x, y) n_j \, dy. \] (9.98)
9.4 Sample results

The homogenized equations (9.26) and (9.58) provide a simple yet powerful tool to investigate the macroscopic elastic dielectric response of deformable dielectrics that, due to their fabrication process, contain from the outset a distribution of space charges in their “ground” state (i.e., in the absence of externally applied electric fields and mechanical forces). As mentioned during the setting of the problem, a prominent example of such a class of materials is electrets (Hilczer and Malecki, 1986; Gerhard-Multhaupt, 1999; Kestelman et al., 2000; Bauer et al., 2004; Hillenbrand and Sessler, 2008; Deng et al., 2014). Similarly, the homogenized equations (9.81) and (9.83) provide a tool to investigate the macroscopic elastic dielectric response of deformable dielectrics that do not contain space charges in their ground state, but that, instead, develop an internal distribution of space charges when externally subjected to an electric field, for instance, by a charge injection process (Lewis, 2004; Roy et al., 2005). Dielectric elastomers filled with (semi-)conducting or high-dielectric nanoparticles have been recently identified as a possible example of such a class of materials (Lewis, 2004; Lopez-Pamies et al., 2014; see also Section 8.5 in Chapter 8). In this section, with the compound purpose of demonstrating the use of the resulting homogenized equations and of illustrating the dominant effect that passive and active charges can have on the macroscopic behavior of elastic dielectrics, we work out two sets of sample results.

9.4.1 A porous electret with passive charges on the walls of the pores

We begin with the results in $N = 1$ space dimension (where all the pertinent calculations can be carried out analytically) for the effective tensors $\tilde{\varepsilon}$, $\tilde{\alpha}$, $\tilde{L}$, $\tilde{M}$, $\tilde{B}^{(1)}$, $\tilde{B}^{(2)}$, $\tilde{B}^{(3)}$ and the macroscopic response under an externally applied uniaxial field of a porous electret made up of alternating layers of an elastic dielectric matrix and air-filled pores bonded through thin interphases that contain passive charges; see, e.g., Hillenbrand and Sessler (2008) and Deng et al. (2014) for experiments and earlier modeling of this problem. From a physical point of view, these results are aimed at demonstrating the use of the homogenized equations (9.39) and (9.75) to describe the well-established piezoelectric-like response of porous electrets due the presence of fixed charges — induced by Corona charging procedures — on the surfaces of the enclosed pores.

Microscopic description of the porous electret We take the matrix phase to be an ideal elastic dielectric with constant permittivity $\varepsilon_m$ and elasticity modulus $\mu_m$. To account for their
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internal pressure (Idiart and Lopez-Pamies, 2012), we also take the air-filled pores to be ideal elastic dielectrics with constant permittivity \(\varepsilon_p\) and elasticity modulus \(\mu_p\) and, moreover, write the sole component (recall that \(N = 1\)) of the local permittivity, elasticity, and electrostriction tensors (9.1) as the scalar functions

\[
\varepsilon_{11}(y) = [1 - \theta_p(y)] \varepsilon_m + \theta_p(y)\varepsilon_p, \quad L_{1111}(y) = 2 [1 - \theta_p(y)] \mu_m + 2\theta_p(y)\mu_p, \quad M_{1111}(y) = \frac{1}{2} \varepsilon_{11}(y),
\]

(9.99)
of the single space variable \(y\) along the Cartesian laboratory axis \(e_1\) aligned with the unit cell \(Y = (0, 1)\); see Fig. 9.5(a) for a schematic. In the above expressions, \(\theta_p(y)\) stands for the indicator function of the spatial regions occupied by the pores and is given by\(^2\)

\[
\theta_p(y) = \begin{cases} 
1 & \text{if } 1 - \frac{c_p}{2} < y < 1 + \frac{c_p}{2} \\
0 & \text{otherwise}
\end{cases}
\]

(9.100)

with \(c_p = \int_Y \theta_p(y)dy\) denoting the volume fraction of pores in the electret. In addition, we model the distribution of passive charges through the choice of constitutive functions

\[
f_1(x) = 1 \quad \text{and} \quad g_1(y) = q[\theta_{i1}(y) - \theta_{i2}(y)]
\]

(9.101)
in (9.10). Here, the parameter \(q\) (of units C/m\(^3\)) stands for the charge density, while

\[
\theta_{i1}(y) = \begin{cases} 
1 & \text{if } 1 - \frac{c_p}{2} < y < \frac{1 - c_p + c_i}{2} \\
0 & \text{otherwise}
\end{cases} \quad \text{and} \quad \theta_{i2}(y) = \begin{cases} 
1 & \text{if } \frac{1 + c_p - c_i}{2} < y < \frac{1 + c_p}{2} \\
0 & \text{otherwise}
\end{cases}
\]

(9.102)

are the indicator functions of the two thin interphasial regions surrounding the pores where the charges are located; see Fig. 9.4(a). In these last expressions, \(c_i = \int_Y \{\theta_{i1}(y) + \theta_{i2}(y)\}dy\) denotes the total volume fraction of the regions containing the charges.

\(^2\)Note that the piecewise constant permittivity (9.99) does not fall within the realm of the regularity (9.5) assumed at the outset. However, as already alluded to in Section 9.1, the homogenization formulae worked out in Section 9.2 and 9.3 remain valid for the type of piecewise constant permittivity considered here and in the next subsection.
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Figure 9.4: (a) Schematic of the unit cell Y illustrating the distribution of pores and the surrounding layers of passive charges within the electret, as characterized by the indicator functions (9.100) and (9.102). (b) Comparison between the macroscopic “piezoelectric” coefficient \( \hat{d} \) (solid line) defined by equation (9.106), experimental data (triangles) of Hillenbrand and Sessler (2008), and an earlier analytical result (dashed line) due to Deng et al. (2014).

The effective coefficients \( \hat{\varepsilon}_{11}, \hat{\alpha}_{11}, \hat{L}_{1111}, \hat{B}_{1111}^{(1)}, \hat{B}_{1111}^{(2)}, \hat{B}_{1111}^{(3)} \). Upon direct use of the local elastic dielectric properties (9.99) and constitutive functions (9.101) characterizing the distribution of charges, the ordinary differential equations that result from (9.22) and (9.55) for the functions \( \omega_1(y), \varphi_1(y), \) and \( \chi_{111}(y) \) can be readily solved in closed form. In turn, the integrals (9.27), (9.28), (9.59), (9.60), and (9.62)–(9.64) can be readily evaluated in closed form to render

\[
\begin{align*}
\hat{\varepsilon}_{11} &= \frac{\varepsilon_n \varepsilon_p}{\varepsilon_n + (1 - c_p) \varepsilon_p}, \\
\hat{\alpha}_{11} &= \frac{c_1 (2c_p - c_1) \varepsilon_n \beta}{4(\varepsilon_p \varepsilon_n + (1 - c_p) \varepsilon_p)}, \\
\hat{L}_{1111} &= \frac{2\mu_n \mu_p}{\varepsilon_n \mu_n + (1 - c_p) \mu_p}, \\
\hat{B}_{1111}^{(1)} &= \frac{1}{4} \left( \frac{1 - c_p}{\varepsilon_n \mu_n} + \frac{c_p}{\varepsilon_p \mu_p} \right) \hat{L}_{1111}, \\
\hat{B}_{1111}^{(2)} &= \frac{1 - c_p}{4\varepsilon_n \mu_n} + \frac{1}{3(2c_p - c_1)^2 \varepsilon_n \mu_p} + \\
&\quad \frac{c_1 (4c_p - 3c_1)(\varepsilon_p \varepsilon_n + 2(1 - c_p) \varepsilon_p)}{12(2c_p - c_1)^2 \varepsilon_n \mu_p} \hat{L}_{1111}.
\end{align*}
\]

For subsequent comparison with some experimental results of Hillenbrand and Sessler (2008), we list in Table 9.1 the values taken by the effective coefficients (9.103) for the choice of material parameters: \( \varepsilon_n = 2.35 \varepsilon_0, \mu_n = 1.0 \text{ GPa}, \varepsilon_p = \varepsilon_0, \mu_p = 0.23 \text{ MPa}, \beta = 0.2 \text{ F/m}, c_p = 0.55, \) and \( c_1 = 0.01; \) recall that \( \varepsilon_0 \approx 8.85 \times 10^{-12} \text{ F/m} \) stands for the permittivity of vacuum. These values corresponds to a polypropylene film with 55% porosity and overall Young’s modulus 0.84 MPa as in the experiments.
of Hillenbrand and Sessler (2008).

<table>
<thead>
<tr>
<th>( \hat{\varepsilon}_{11} )</th>
<th>( \hat{\alpha}_{11} ) (F/m)</th>
<th>( \hat{L}_{1111} ) (MPa)</th>
<th>( \hat{M}_{1111} )</th>
<th>( \hat{B}^{(1)}_{1111} ) (F/m)</th>
<th>( \hat{B}^{(3)}_{1111} ) (F/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.35( \varepsilon_0 )</td>
<td>7.35 \times 10^{-4}</td>
<td>0.85</td>
<td>0.91( \varepsilon_0 )</td>
<td>-1.72 \times 10^{-4}</td>
<td>4.04 \times 10^3</td>
</tr>
</tbody>
</table>

Table 9.1: Computed values of the seven effective coefficients \( \hat{\varepsilon}_{11}, \hat{\alpha}_{11}, \hat{L}_{1111}, \hat{M}_{1111}, \hat{B}^{(1)}_{1111}, \hat{B}^{(2)}_{1111}, \hat{B}^{(3)}_{1111} \) characterizing the macroscopic properties of the porous electret with charges (\( \beta = 0.2 \) F/m) on the walls of the enclosed pores.

### Macroscopic response of a thin film under a uniaxial electric field

Now that the seven effective coefficients (9.103) have been determined, any boundary-value problem of interest may be investigated with help of the homogenized equations (9.26) and (9.58). Here, we consider a popular one in experiments wherein a thin film of thickness \( t \) made up of the porous electret is subjected to a uniaxial electric field across its thickness through the application of a voltage \( \Phi \). In such a setup, neglecting fringe effects, the governing equations (9.26) and (9.58) are trivially satisfied and the macroscopic electric potential and macroscopic displacement field are given (up to an additive constant) by

\[
\varphi(x) = -E_1 x \quad \text{and} \quad u_1(x) = \overline{\varphi}_{11} x
\]

with

\[
E_1 = -\frac{\Phi}{t} \quad \text{and} \quad \overline{\varphi}_{11} = -\frac{\hat{M}_{1111} E_1^2}{L_{1111}} - \frac{2\hat{B}^{(1)}_{1111}}{L_{1111}} E_1 - \frac{\hat{B}^{(3)}_{1111}}{L_{1111}}.
\]

(9.105)

A quantity of significant practical interest that can be readily extracted from the solution (9.104)–(9.105) is the macroscopic “piezoelectric” coefficient

\[
d = \frac{\partial \overline{\varphi}_{11}}{\partial E_1} = -\frac{2\hat{M}_{1111}}{L_{1111}} E_1 - \frac{2\hat{B}^{(1)}_{1111}}{L_{1111}}
\]

\[
= -\frac{1}{2} \left( \frac{1}{\varepsilon_n \mu_n} + \frac{c_p}{\varepsilon_p \mu_p} \right) \hat{\varepsilon}_{11} E_1 - \frac{(1 - c_p)(\mu_p - \mu_n)}{2\varepsilon_n \mu_n \mu_p} \hat{\varepsilon}_{11} \hat{\alpha}_{11}.
\]

(9.106)

For comparison with the experimental data (triangles) of Hillenbrand and Sessler (2008) for a 71 \( \mu \)m-thick polypropylene film with 55% porosity, this coefficient is plotted (solid line) in Fig. 6.8, as a function of the applied electric field \( E_1 \), for the numerical values of the parameters listed in Table 9.2. For further comparisons, the earlier analytical result (dashed line) of Deng et al. (2014) is also included in the figure.

---

\( \text{Note that the pores in the specimens of Hillenbrand and Sessler (2008) were of oblate spheroidal shape, and not exactly layers as in the sample calculations presented here.} \)
9.4.2 A dielectric elastomer composite with interphasial active charges

In the sequel, we present sample results for \( N = 3 \) (i.e., in three spatial dimensions) for the effective tensors \( \tilde{\varepsilon}, \tilde{L}, \tilde{M} \) and the macroscopic electrostriction response of a dielectric elastomer composite made up of an elastic dielectric matrix filled with firmly bonded particles surrounded by a thin interphasial layer of active charges. The results are aimed at demonstrating the use of the homogenized equations (9.81) and (9.83), at illustrating the dominant effect that space charges can have on the macroscopic behavior, as well as at supporting the conjecture that the extreme enhancement of the macroscopic elastic dielectric response of emerging dielectric elastomer composites observed in recent experiments (see, e.g., Huang et al., 2005; Liu et al., 2013) is due to the presence of space charges surrounding the underlying filler particles.

Microscopic description of the dielectric elastomer composite For definiteness, we consider the case when both the matrix and the particles are ideal elastic dielectrics so that the permittivity, elasticity, and electrostriction tensors (9.1) can be written in terms of a scalar permittivity function

\[
\varepsilon(y) = [1 - \theta_p(y)] \varepsilon_m + \theta_p(y) \varepsilon_p, \tag{9.107}
\]
a shear modulus function

\[
\mu(y) = [1 - \theta_p(y)] \mu_m + \theta_p(y) \mu_p, \tag{9.108}
\]
and a bulk modulus function

\[
\kappa(y) = [1 - \theta_p(y)] \kappa_m + \theta_p(y) \kappa_p, \tag{9.109}
\]
rather simply as (see, e.g., Chapters 3 and 4)

\[
\varepsilon_{ij}(y) = \varepsilon(y) \delta_{ij}, \quad L_{ijkl}(y) = 2\mu(y)\mathcal{K}_{ijkl} + 3\kappa(y)\mathcal{J}_{ijkl}, \quad M_{ijkl}(y) = \varepsilon(y)\mathcal{K}_{ijkl} - \frac{\varepsilon(y)}{2}\mathcal{J}_{ijkl} \tag{9.110}
\]

with

\[
\mathcal{K}_{ijkl} = \frac{1}{2} \left( \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl} \right) \quad \text{and} \quad \mathcal{J}_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl}. \tag{9.111}
\]

In the above expressions, \( \varepsilon_m, \mu_m, \kappa_m \), and \( \varepsilon_p, \mu_p, \kappa_p \) denote the constant permittivity\(^4\), shear modulus, and bulk modulus of the matrix and the particles, while \( \theta_p(y) \) denotes the indicator function of the

\(^4\)Note that the piecewise constant permittivity (9.107) does not fall within the realm of the regularity (9.5) assumed at the outset. However, as already alluded to in Section 9.1, the homogenization formulae worked out in Sections 9.2 and 9.3 remain valid for the type of piecewise constant permittivity considered here.
spatial regions occupied by the particles. Here, the focus is on filler particles that are spherical in shape, all of the same size, and that are distributed in a cubic array. Specifically, in our calculations, we set

\[
\theta_p(y) = \begin{cases} 
1 & \text{if } |y - y_p| < \left( \frac{3c_p}{4\pi} \right)^{1/3}, \quad y_p = \left( \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \right), \\
0 & \text{otherwise}
\end{cases}
\tag{9.112}
\]

where \(c_p = \int_Y \theta_p(y) dy\) stands for the volume fraction of particles in the composite. Throughout this section, the components of all tensorial quantities are referred to the Cartesian laboratory axes \(e_1, e_2, e_3\) aligned with the unit cell \(Y = (0, 1)^3\), as depicted in Fig. 9.5(a).

Moreover, following Lopez-Pamies et al. (2014), we take the space-charge density function (9.10) to be characterized (recall that for the case of active charges the constitutive function \(f(x)\) is prescribed as \(f_i(x) = -\partial \varphi(x)/\partial x_i\)) by the function

\[
g(y) = \theta_\bar{1}(y) \beta \frac{y - y_p}{|y - y_p|}, \tag{9.113}
\]

where \(\beta\) is a constant of choice (of units F/m) that physically can be viewed as a measure of charge content, while \(\theta_\bar{1}(y)\) stands for the indicator function of the spatial regions wherein the charges are contained. Here, the focus is on space charges that are contained within an interphasial layer of constant thickness surrounding the particles. Specifically, we set

\[
\theta_\bar{1}(y) = \begin{cases} 
1 & \text{if } \left( \frac{3c_p}{4\pi} \right)^{1/3} < |y - y_p| < \left( \frac{3}{4\pi} \left( c_p + c_\bar{1} \right) \right)^{1/3}, \\
0 & \text{otherwise}
\end{cases}
\tag{9.114}
\]

where \(y_p\) in (9.113)–(9.114) is as in expression (9.112) and \(c_\bar{1} = \int_Y \theta_\bar{1}(y) dy\) denotes the volume fraction of the spatial regions that contain the charges. Figure 9.5(a) shows a schematic of the defining unit cell \(Y\) for the above-described dielectric elastomer composite.
Figure 9.5: (a) Schematic of the unit cell $Y$ illustrating the cubic distribution of spherical particles and the surrounding layer of space charges, as characterized by the indicator functions (9.112) and (9.114). (b) Finite element discretization (with a total of about 760,000 tetrahedral elements) of the unit cell $Y$ utilized to solved numerically the pdes (9.89)–(9.55) for the functions $\hat{\omega}_i(y)$ and $\chi_{ijk}(y)$ required in the calculations of the effective tensors $\tilde{\varepsilon}$, $\tilde{\mathbf{L}}$, and $\tilde{\mathbf{M}}$. Both parts (a) and (b) correspond to the same case of a volume fraction of filler particles $c_p = 0.073$ and a volume fraction of surrounding interphases $c_i = 0.038$.

The effective permittivity, elasticity, and electrostriction tensors $\tilde{\varepsilon}$, $\tilde{\mathbf{L}}$, and $\tilde{\mathbf{M}}$ Upon direct use of the local elastic dielectric properties (9.110) and the constitutive function (9.113) for the space charges, the formula (9.82) for the effective permittivity tensor specializes to

$$\tilde{\varepsilon}_{ij} = \int_Y \varepsilon(y) \left( \delta_{ij} + \frac{\partial \hat{\omega}_i(y)}{\partial y_j} \right) dy + \int_Y \theta_i(y) \frac{y_j - y_{ij}}{|y - y_p|} dy,$$  \hspace{1cm} (9.115)

while the formulæ (9.59) and (9.84) for the effective elasticity tensor and the effective electrostriction tensor specialize to

$$\tilde{\mathbf{L}}_{ijkl} = \int_Y 2\mu(y) K_{ijpq} \left( \delta_{pk} \delta_{qk} + \frac{\partial \chi_{pq}}{\partial y_q} (y) \right) dy + \int_Y \kappa(y) \delta_{ij} \left( \delta_{kl} + \frac{\partial \chi_{kl}}{\partial y_p} (y) \right) dy$$  \hspace{1cm} (9.116)

and

$$\tilde{\mathbf{M}}_{ijkl} = \int_Y \varepsilon(y) K_{rspq} \left( \delta_{rt} \delta_{s} + \frac{\partial \chi_{rij}}{\partial y_i} (y) \right) \left( \delta_{pk} + \frac{\partial \hat{\omega}_k}{\partial y_p} (y) \right) \left( \delta_{ql} + \frac{\partial \hat{\omega}_l}{\partial y_q} (y) \right) dy - \frac{1}{6} \int_Y \varepsilon(y) \left( \delta_{ij} + \frac{\partial \chi_{ij}}{\partial y_r} (y) \right) \left( \delta_{jq} + \frac{\partial \hat{\omega}_q}{\partial y_r} (y) \right) \left( \delta_{il} + \frac{\partial \hat{\omega}_l}{\partial y_q} (y) \right) dy.$$  \hspace{1cm} (9.117)

As discussed in remark iv of Section 9.4, for given values of the permittivity, shear modulus, and bulk modulus $\varepsilon_m$, $\mu_m$, $\kappa_m$, and $\varepsilon_p$, $\mu_p$, $\kappa_p$ of the matrix and filler particles, given value of the space-charge content parameter $\beta$, and given values of the volume fractions $c_p$ and $c_i$ of the particles and interphases, the linear elliptic pdes (9.89)–(9.55) for $\hat{\omega}_i(y)$ and $\chi_{ijk}(y)$ can be readily solved numerically, for instance, by the finite element method. In turn, the integrals (9.115)–(9.117) can be evaluated by means of a quadrature rule to determine the effective tensors $\tilde{\varepsilon}$, $\tilde{\mathbf{L}}$, and $\tilde{\mathbf{M}}$. 
For later comparison with an experiment of Huang et al. (2005), we work out here the solutions for the following two sets of values of the various parameters: \( \varepsilon_m = 8 \varepsilon_0 \), \( \mu_m = 0.819 \) MPa, \( \kappa_m = 1 \) GPa, \( \varepsilon_p = 10^4 \varepsilon_0 \), \( \mu_p = 1 \) GPa, \( \kappa_p = 1 \) GPa, \( \beta = 0 \) F/m and 6500 \( \varepsilon_0 \), \( c_p = 0.073 \), and \( c_1 = 0.038 \); recall that \( \varepsilon_0 \approx 8.85 \times 10^{-12} \) F/m stands for the permittivity of vacuum. These values correspond to a polyurethane matrix filled with 7.3% volume fraction of particles made up of the semi-conducting polymer o-CuPc, as in the experiments of Huang et al. (2005). For the case with \( \beta = 6500 \varepsilon_0 \), the o-CuPc particles are surrounded by a thin interphasial layer of space charges occupying a total of 3.8% volume fraction of the composite. For the case with \( \beta = 0 \) F/m, on the other hand, the composite does not contain any space charges. For both sets of calculations, with and without charges (\( \beta = 6500 \varepsilon_0 \) and 0 F/m), we solve the pdes (9.89)--(9.55) for \( \tilde{\omega}_i(y) \) and \( \chi_{ijk}(y) \) by means of the finite element method. We make use in particular of the finite dimensional subspaces and approach presented in Section 5.1; Fig. 9.5(b) depicts the finite element discretization of the unit cell \( Y \) employed here to carry out the pertinent calculations, which was checked to be sufficiently refined to lead to accurate solutions.

Now, owing to the spherical shape and cubic spatial distribution of the particles, the constitutive isotropy of their elastic dielectric properties (9.110), and the form (9.113) with (9.114) for the function \( g(y) \) characterizing the distribution of space charges, it follows that the resulting effective permittivity tensor (9.115) is an isotropic tensor, while the resulting effective elasticity tensor (9.116) and the effective electrostriction tensor (9.117) possess cubic symmetry. With help of the Walpole notation (Walpole, 1981)

\[
S_{ijkl} = \begin{cases} 
1 & \text{if } i = j = k = l \\
0 & \text{otherwise}
\end{cases}, \quad K'_{ijkl} = S_{ijkl} - J_{ijkl}, \quad K''_{ijkl} = K_{ijkl} - K'_{ijkl}, \quad (9.118)
\]

they can be expediently written as

\[
\tilde{\varepsilon}_{ij} = \varepsilon \delta_{ij}, \quad \tilde{L}_{ijkl} = 2\tilde{\mu}'K'_{ijkl} + 2\tilde{\mu}''K''_{ijkl} + 3\kappa J_{ijkl}, \quad \tilde{M}_{ijkl} = m'_K K'_{ijkl} + m''_K K''_{ijkl} + m_J J_{ijkl}. \quad (9.119)
\]

Table 9.2 presents the results generated numerically for all seven effective material coefficients in (9.119) for the dielectric elastomer composite with (\( \beta = 6500 \varepsilon_0 \)) and without (\( \beta = 0 \) F/m) interphasial charges. From a physical point of view, as already discussed in remark ii of Section 9.4, the key point to recognize from the sample results in Table 9.2 is that suitable distributions of space charges can lead to extremely large values (in principle, positive or negative as large as desired) of the effective permittivity and electrostriction coefficients of dielectric elastomer composites.
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Table 9.2: Computed values of the seven effective material coefficients \( \tilde{\varepsilon}, \tilde{\mu}', \tilde{\mu}'', \tilde{\kappa}, \tilde{m}', \tilde{m}'', \tilde{m} \) in the effective permittivity, elasticity, and electrostriction tensors (9.119) characterizing the macroscopic properties of the dielectric elastomer with \( (\beta = 6500 \varepsilon_0) \) and without \( (\beta = 0 \text{ F/m}) \) interphasial charges.

<table>
<thead>
<tr>
<th>( \beta )</th>
<th>( \tilde{\varepsilon} )</th>
<th>( \tilde{\mu}' ) (MPa)</th>
<th>( \tilde{\mu}'' ) (MPa)</th>
<th>( \tilde{\kappa} ) (GPa)</th>
<th>( \tilde{m}'_K )</th>
<th>( \tilde{m}''_K )</th>
<th>( \tilde{m}_J )</th>
</tr>
</thead>
<tbody>
<tr>
<td>6500 ( \varepsilon_0 )</td>
<td>29.1 ( \varepsilon_0 )</td>
<td>1.01</td>
<td>0.96</td>
<td>1.00</td>
<td>207 ( \varepsilon_0 )</td>
<td>134 ( \varepsilon_0 )</td>
<td>-235 ( \varepsilon_0 )</td>
</tr>
<tr>
<td>0 \text{ F/m}</td>
<td>9.89 ( \varepsilon_0 )</td>
<td>1.01</td>
<td>0.96</td>
<td>1.00</td>
<td>10.2 ( \varepsilon_0 )</td>
<td>9.80 ( \varepsilon_0 )</td>
<td>-4.94 ( \varepsilon_0 )</td>
</tr>
</tbody>
</table>

Macroscopic electrostriction response

Having determined the effective permittivity, elasticity, and electrostriction tensors \( \tilde{\varepsilon}, \tilde{\mu}', \tilde{\mu}'', \tilde{\kappa}, \tilde{m}', \tilde{m}'', \tilde{m} \), the homogenized equations (9.81) and (9.83) can now be utilized to study any boundary-value problem of interest. Here, we shall study a simple one that is pervasive in experiments: a thin layer of dielectric elastomer composite that is sandwiched between two compliant electrodes connected to a battery, as depicted schematically in Fig. 9.6(a). The deformation induced in the composite by the applied voltage in such a setting is referred to as electrostriction.

Specifically, we consider a layer specimen whose thickness \( d_3 \) is much smaller than its other two dimensions \( (d_3 \ll d_1, d_2) \), so that fringe effects can be neglected. Further, we take the geometry of the specimen to be co-axial with the underlying cubic distribution of the filler particles and the voltage \( \Phi \) applied across the compliant electrodes to be in the \( e_3 \) direction; see Fig. 9.6(a). In such a configuration, the homogenized equations (9.81) and (9.83) happen to be satisfied trivially, in particular, the macroscopic electric potential and macroscopic displacement field are given (up to
an additive constant) by the affine relations

\[ \varphi(x) = -E_i x_i \quad \text{and} \quad u_i(x) = H_{ij} x_j, \]  

(9.120)

where

\[ E_1 = E_2 = 0, \quad E_3 = -\frac{\Phi}{d_3}, \]  

(9.121)

and

\[ \begin{align*}
H_{11} &= H_{22} = \left( \frac{m_K'}{6\mu'} - \frac{m_J}{9\kappa} \right) E_3^2, \\
H_{33} &= -\left( \frac{m_K'}{3\mu'} + \frac{m_J}{9\kappa} \right) E_3^2, \\
H_{12} &= H_{21} = H_{13} = H_{31} = H_{23} = H_{32} = 0. 
\end{align*} \]  

(9.122)

The electrostriction strain \( \overline{H}_{33} \) in the direction of the applied voltage is plotted in Fig. 9.6(b) as a function of the corresponding electric field \( E_3 \) for the composite with charges (solid line) and without charges (dashed line). For comparison purposes, the experimental data (solid triangles) of Huang et al. (2005) for a polyurethane elastomer filled with roughly spherical o-CuPc particles at volume fraction \( c_p = 0.073 \) is also included in the figure. From a physical point of view, the key feature to recognize from the results in Fig. 9.6(b) is that they are supportive of the viewpoint that the extreme macroscopic elastic dielectric properties of emerging dielectric elastomer composites are due to the presence of space charges surrounding the underlying filler particles; see Section 8.5 in Chapter 8 for related calculations for isotropic composites — in the context of finite deformations and finite magnetic fields — providing further supporting evidence.

---

5Note that the filler particles in the specimens of Huang et al. (2005) were distributed isotropically, and not periodically in a cubic array as in the sample calculations presented here.
A general result for the magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber, with applications to spherical and cylindrical specimens

Case I Trivial statement
If an elegant theory agrees with experiment, there is nothing to worry about.

Case II Heisenberg’s postulate
If an elegant theory does not agree with experiment, the experiment must be wrong.

Case III Bohr’s amendment
If an inelegant theory disagrees with experiment, the case is not lost because [by] improving the theory one can make it agree with experiment.

Case IV My opinion
If an inelegant theory agrees with experiment, the case is hopeless.

– George Gamow, in a letter to Paul Dirac
The present chapter is concerned with the homogenized (or macroscopic) magnetoelastic response of magnetorheological elastomers comprised of non-Gaussian rubbers filled with isotropic suspensions of either iron or ferrofluid particles. Leveraging and extending the results in Chapters 7 and 8 within the mathematically analogous setting of electroelastostatics, we put forth a homogenization-based macroscopic free energy that describes the finite magnetoelastic response of isotropic magnetorheological elastomers under arbitrary magnetomechanical loadings. The focus is on isotropic magnetorheological elastomers — in both $N = 2$ and 3 space dimensions — comprised of a non-Gaussian rubber matrix isotropically filled with either iron or ferrofluid particles; see Fig. 10.1 for a schematic. By deploying the constructed free energies, we provide insight into the merits of using ferrofluid particles in lieu of the more conventional iron particles as fillers and scrutinize experiments available in the literature on magnetorheological elastomers containing iron particles. This is accomplished by carrying out finite-element simulations of representative experiments making use of the constructed free energies to model the magnetoelastic behavior of the specimens.

To put the present chapter in perspective, we remark that due to the renewed experimental impetus started during the 1990s (see, e.g., Shiga et al., 1995; Jolly et al., 1996; Ginder et al., 1999), increasing efforts have been devoted by the mechanics community to construct continuum models capable of describing the magnetoelastic response of magnetorheological elastomers under finite deformations (involving arbitrary finite strains and rigid rotations) and finite magnetic fields. These efforts can be roughly classified into two categories: (i) top-down or phenomenological approaches in which macroscopic free energies are postulated based on macroscopic experimental observations (see,
e.g., Kankanala and Triantafyllidis, 2004; Dorfmann and Ogden, 2005b; Bustamante et al., 2011; Danas et al., 2012; Saxena et al., 2015; Pelteret et al., 2016) and (ii) bottom-up or homogenization approaches in which macroscopic free energies are derived based on the underlying microscopic behavior (see, e.g., Borcea and Bruno, 2001; Zhou and Shin, 2005; Liu et al., 2006; Corcolle et al., 2008; Galipeau and Ponte Castañeda, 2012, 2013). While the practical challenges of carrying out experiments that test the material (and not the structural) response of specimens over wide ranges of finite deformations and finite magnetic fields have curtailed the advancement of phenomenological models, the intrinsic mathematical challenges of carrying out the homogenization limit of the equations of magnetoelastostatics have hindered the construction of homogenization-based models.

Consequently, existing analytical homogenization (exact or approximate) results for isotropic magnetorheological elastomers are restricted to the asymptotic context of small deformations, save for an approximate result due to Galipeau and Ponte Castañeda (2012) in $N = 2$ space dimensions that is valid for finite deformations. These authors made use of a partial decoupling approximation (Ponte Castañeda and Galipeau, 2011) together with an earlier result of Lopez-Pamies and Ponte Castañeda (2006) to construct an estimate for the macroscopic free energy of an isotropic incompressible elastic matrix reinforced by an isotropic suspension of circular magnetizable particles that are mechanically rigid. It is also fitting to remark that computational homogenization results have been recently reported in the literature for rubber filled with periodic square/hexagonal arrays (Javili et al., 2013; Galipeau et al., 2014; Keip and Rambausek, 2016) and approximately isotropic distributions (Kalina et al., 2016) of circular particles in $N = 2$ space dimensions and with periodic cubic arrays of spherical particles (Javili et al., 2013; Miehe et al., 2016) in $N = 3$ space dimensions. These computational results pertain to rubber matrices featuring uncharacteristically high compressibility (presumably in order to avoid numerical complications such as volumetric locking).

We also remark that neither theoretical nor experimental studies on magnetorheological elastomers containing ferrofluid filler particles appear to have been reported in the literature; see, however, the results in Chapter 8, the recent works of Lopez-Pamies (2014), Barlett et al. (2017), and references therein for intimately related studies of dielectric elastomers filled with liquid-metal inclusions.
10. Magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber

10.1 The problem

Microscopic description of the material  We are interested in describing the macroscopic magnetoelastic response of a rubber matrix filled with a statistically uniform and isotropic suspension of firmly bonded iron or ferrofluid particles under finite deformations and finite magnetic fields. This so-called magnetorheological elastomer is taken to occupy a $N$-dimensional domain $\Omega \subset \mathbb{R}^N$ ($N = 2, 3$), with boundary $\partial \Omega$, in its undeformed, stress-free, and magnetization-free configuration; for convenience, we choose units of length so that $|\Omega| = 1$. The rubber matrix occupies a domain $\Omega_m$, while the particles — which are taken to be of much smaller sizes than the macroscopic length scale — occupy collectively its complement $\Omega_p = \Omega \setminus \Omega_m$; see Fig. 10.1.

Each material point in the ground configuration $\Omega$ is identified by its initial position vector $X$, while its position in the deformed configuration $\omega$ is given by $x = \chi(X)$. We assume that the mapping $\chi$ is bijective, continuous, and sufficiently regular to warrant the mathematical well-posedness of the equations that follow. The corresponding deformation gradient is denoted by $F = \text{Grad} \chi$.

The constitutive behaviors of the matrix and filler particles are taken to be characterized by “total” free-energy functions (Dorfmann and Ogden, 2004) of the deformation gradient $F$ and Lagrangian magnetic field $H$, in particular, of the $(I_1, I_H)$–based form

$$W_m(F, H) = \begin{cases} \Psi(I_1) - \frac{\mu_0}{2} I_H & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \quad (10.1)$$

and

$$W_p(F, H) = \begin{cases} \frac{G_p}{2} [I_1 - N] - S(I_H) & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \quad (10.2)$$

In these expressions, $I_1 = F \cdot F$, $J = \det F$, $I_H = F^{-T} H \cdot F^{-T} H$, $\mu_0 = 4\pi \times 10^{-7} \text{ H/m}$ is the permeability of vacuum, $G_p$ stands for the initial shear modulus of the particles, $\Psi$ denotes any non-negative function of choice (suitably well-behaved) satisfying the linearization conditions $\Psi(N) = 0$, $\Psi'(N) = G/2$ with $G$ denoting the initial shear modulus of the rubber, and the function $S$ is also a function of choice satisfying the linearization conditions $S(0) = 0$, $S'(0) = \mu_p/2$ and the convexity conditions $S'(I_H) > 0$, $S'(I_H) + 2I_H S''(I_H) > 0$, where $\mu_p$ stands for the initial permeability of the particles.

---

1 By considering the cases $N = 2$ and $N = 3$ simultaneously, we are able to deal at the same time with suspensions of (i) aligned cylindrical fibers and (ii) three-dimensional particles. In both cases, we shall refer to the iron or ferrofluid fillers as particles.

2 We recall here the use of the standard convention $y'(x) = dy(x)/dx$ to denote the derivative of functions of a single scalar variable.
Given the free-energy functions (10.1) and (10.2), it follows that the first Piola-Kirchhoff stress tensor $S$ and Lagrangian magnetic induction $B$ at any material point $X \in \Omega$ are given expediently by the relations

$$S(X) = \frac{\partial W}{\partial F}(X, F, H) \quad \text{and} \quad B(X) = -\frac{\partial W}{\partial H}(X, F, H)$$

(10.3)

with

$$W(X, F, H) = [1 - \theta_p(X)]W_n(F, H) + \theta_p(X)W_p(F, H),$$

(10.4)

where $\theta_p(X)$ is the characteristic function of $\Omega_p$: $\theta_p(X) = 1$ if $X \in \Omega_p$ and zero otherwise. It further follows that the total Cauchy stress $T$, Eulerian magnetic induction $b$, and magnetization $m$ (per unit deformed volume) are in turn given by $T = SF$, $b = FB$, and $m = \mu^{-1}b - h$ with $h = F^{-T}H$ denoting the Eulerian magnetic field. We note that the built-in material frame indifference of (10.1)–(10.2) ensures that $T^T = T$.

Before proceeding with the description of the macroscopic response of the above-defined magneto-rheological elastomer, we remark that free-energy functions of the form (10.1) have been shown to describe reasonably well the response of a broad variety of rubbers — which are intrinsically non-magnetizable — over wide ranges of deformations (see, e.g., Gent, 1996; Lopez-Pamies, 2010; Nunes and Moreira, 2013; Ritto and Nunes, 2015). While an analytical result will be presented in Section 10.2 that is valid for arbitrary choices of the function $\Psi$, sample numerical results will be presented in Sections 10.3 through 10.6 for the choice

$$\Psi(I_1) = \frac{N^{1-\alpha_1}}{2\alpha_1}G_1[I_1^{\alpha_1} - N^{\alpha_1}] + \frac{N^{1-\alpha_2}}{2\alpha_2}G_2[I_1^{\alpha_2} - N^{\alpha_2}].$$

(10.5)

In this expression, we recall that $N$ stands for the space dimension ($N = 2, 3$) and $G_1, G_2, \alpha_1, \alpha_2$ are real-valued material parameters that may be associated with the non-Gaussian statistical distribution of the underlying polymer chains. In addition to its mathematical simplicity and physical meaning of its parameters, we choose this class of functions because of its rich functional form and demonstrated descriptive and predictive capabilities (Lopez-Pamies, 2010).

Moreover, free-energy functions of the form (10.2) are expected to describe reasonably well the finite magnetoelastic response of a spectrum of magnetizable filler particles ranging from carbonyl iron to ferrofluids; while carbonyl iron has already been widely utilized as filler particles by the experimental community, the authors are not aware of experiments involving ferrofluid filler particles (see, however, the device explored by Wang and Gordaninejad, 2009). We emphasize in particular
that free-energy functions of the form (10.2) are general enough to model (albeit ignoring dissipative effects) magnetization saturation phenomena (see, e.g., Arias et al., 2006; Ivanov et al., 2007). In this case, granted that the magnetization of the particles is given by

\[ m_p = \left[ \frac{2}{\mu_0} S'(I_5^H) - 1 \right] \mathbf{F}^{-T} \mathbf{H}, \quad (10.6) \]

it must be required, in addition to the linearization and convexity conditions on \( S \) mentioned above, that

\[ S'(I_5^H) = \frac{\mu_0}{2} + \frac{\mu_0 m_s}{2 \sqrt{I_5^H}} + o \left( \frac{1}{\sqrt{I_5^H}} \right) \quad (10.7) \]

in the limit as \( I_5^H \to \infty \). Here, the positive material constant \( m_s \) characterizes the magnitude of the saturated magnetization. While an analytical result will be presented in Section 10.2 that is valid for any function \( S \) of choice, in Sections 10.3 through 10.6 sample numerical results will be presented for the Langevin-type function

\[ S(I_5^H) = \frac{\mu_0}{2} I_5^H + \frac{\mu_0 m_s}{\beta} \ln \left( \frac{\sinh \left( \beta \sqrt{I_5^H} \right)}{\beta \sqrt{I_5^H}} \right) \quad (10.8) \]

where \( \beta = 3(\mu_p - \mu_0)/(\mu_0 m_s) \), so that

\[ m_p = \frac{m_s}{\sqrt{I_5^H}} \left[ \coth \left( \beta \sqrt{I_5^H} \right) - \frac{1}{\beta \sqrt{I_5^H}} \right] \mathbf{F}^{-T} \mathbf{H}. \quad (10.9) \]

**The macroscopic response** In light of the assumed separation of length scales and statistical uniformity of the microstructure, the *microscopically* heterogeneous magnetorheological elastomer described above is expected to behave *macroscopically* as a homogeneous material. Its macroscopic or overall magnetoelastic response can be defined by the relation between the volume averages of the first Piola-Kirchhoff stress \( S \) and Lagrangian magnetic induction \( B \) and the volume averages of the deformation gradient \( \mathbf{F} \) and Lagrangian magnetic field \( \mathbf{H} \) over \( \Omega \) when subjected to the affine boundary conditions \( \mathbf{x} = \mathbf{F} \mathbf{X} \) and \( \psi = -\mathbf{H} \cdot \mathbf{X} \) on \( \partial \Omega \), where the second-order tensor \( \mathbf{F} \) and vector \( \mathbf{H} \) are prescribed quantities\(^3\). Thanks to the identities \( \int_\Omega \mathbf{F} \mathbf{X} \, d\mathbf{X} = \mathbf{F} \) and \( \int_\Omega \mathbf{H} \mathbf{X} \, d\mathbf{X} = \mathbf{H} \) that follow from the divergence theorem, the derivation of the macroscopic response reduces then to computing the average Piola-Kirchhoff stress \( \mathbf{S} = \int_\Omega S \mathbf{X} \, d\mathbf{X} \) and average Lagrangian magnetic induction \( \mathbf{B} = \int_\Omega B \mathbf{X} \, d\mathbf{X} \) in terms of \( \mathbf{F} \) and \( \mathbf{H} \). These macroscopic constitutive relations can be

\(^3\)Here, we have made use of Ampère’s law and represented \( \mathbf{H} \) as the gradient of a scalar potential, namely, \( \mathbf{H} = -\nabla \psi \).
conveniently written in the variational form (Ponte Castañeda and Galipeau, 2011; Lopez-Pamies, 2014)

\[ S = \frac{\partial W}{\partial F}(F, H) \quad \text{and} \quad B = -\frac{\partial W}{\partial H}(F, H), \quad (10.10) \]

where

\[ W(F, H) = \min_{F \in \mathcal{K}} \max_{H \in \mathcal{H}} \int_{\Omega} W(X, F, H) \, dX \quad (10.11) \]

denotes the effective free-energy function of the magnetorheological elastomer; in the above expression, \( \mathcal{K} \) and \( \mathcal{H} \) stand for sufficiently large functional spaces of deformations gradients \( F \) and curl-free magnetic fields \( H \) that are consistent with the applied affine boundary conditions.

In the present context of magnetoelastostatics, we remark that two of the four relevant governing equations, namely, balance of linear momentum and Gauss's law for magnetism,

\[ \text{Div} \, S(X) = 0 \quad \text{and} \quad \text{Div} \, B(X) = 0, \quad (10.12) \]

correspond to the Euler-Lagrange equations associated with the variational problem (10.11). On the other hand, balance of angular momentum is guaranteed from the material frame indifference of the free-energy functions (10.1)–(10.2), while the choice of admissible curl-free magnetic fields \( H \) in the variational problem (10.11) ensures that Ampère's law is satisfied.

In analogy with the above relations between the local Lagrangian and Eulerian quantities, it is not difficult to show that \( \mathbf{T} = S F^T \), \( \mathbf{b} = F \mathbf{B} \), and \( \mathbf{m} = \mu_0^{-1} \mathbf{b} - \mathbf{h} \), where \( \mathbf{T} = |\omega|^{-1} \int_\omega T(x) \, dx \), \( \mathbf{b} = |\omega|^{-1} \int_\omega b(x) \, dx \), \( \mathbf{m} = |\omega|^{-1} \int_\omega m(x) \, dx \) are the volume averages of the Cauchy stress \( T \), Eulerian magnetic induction \( b \), and magnetization \( m \) over the deformed configuration \( \omega \), while \( \mathbf{h} = F^{-T} H \) corresponds to the volume average of the Eulerian magnetic field \( h \) over \( \omega \).

**Isotropic magnetorheological elastomers**  

Granted the assumed isotropy of the microstructure and the constitutive isotropy and incompressibility of the rubber and particles, the macroscopic magnetoelastic response of the magnetorheological elastomer is itself isotropic and incompressible. As a result, its effective free energy (10.11) only depends on the macroscopic deformation gradient \( \mathbf{F} \) and macroscopic Lagrangian magnetic field \( \mathbf{H} \) through \( 2N - 1 \) independent invariants and becomes unbounded for non-isochoric deformations when \( J = \det \mathbf{F} \neq 1 \). With a slight abuse of notation, we shall write for \( N = 2 \)

\[ W(F, H) = \begin{cases} W(I_1, I_4^H, I_5^H) & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases}, \quad (10.13) \]
10. Magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber

and for $N = 3$

$$W(F, H) = \begin{cases} W(T_1, T_2, T_4^H, T_5^H, T_6^H) & \text{if } J = 1, \\ +\infty & \text{otherwise}, \end{cases} \quad (10.14)$$

in terms of the standard invariants

$$T_1 = F \cdot F, \quad T_2 = F^{-T} \cdot F^{-T},$$

$$T_4^H = H \cdot H, \quad T_5^H = F^{-T} H \cdot F^{-T} H, \quad T_6^H = F^{-1} F^{-T} H \cdot F^{-1} F^{-T} H. \quad (10.15)$$

Note that for $N = 2$ we have the connections $T_2 = T_1$ and $T_6^H = T_1 T_5^H - T_4^H$.

10.2 An approximate closed-form solution

We put forth in this section a variational solution for the effective free-energy function $W$ defined by the problem (10.11). To this end, exploiting the well-known mathematical analogy between electroelastostatics and magnetoelastostatics (see, e.g., Stratton, 1941), we invoke the solution (8.47) derived in Chapter 8 for the analogous problem of the nonlinear electroelastic deformation of dielectric elastomer composites in $N = 3$ space dimensions and recast it mutatis mutandis — as well as extend it to $N = 2$ — for the nonlinear magnetoelastic deformation of the magnetorheological elastomers of interest in this chapter. While the general solution (8.47) derived in Chapter 8 applies to deformable particles with arbitrary initial shear modulus $G_p \geq 0$, we restrict the exposition here to the limiting cases of rigid ($G_p = +\infty$) and liquid ($G_p = 0$) particles, which provide reasonable approximations\(^4\) for the iron and ferrofluids fillers of interest here.

The solution Thus, the effective free-energy function (10.11) for a rubber with free-energy function (10.1), filled with any type of non-percolative isotropic suspension of rigid ($G_p = +\infty$) or liquid ($G_p = 0$) particles with free-energy function (10.2) at volume fraction $c$, is given by

$$W(F, H) = \begin{cases} (1 - c)\Psi(I_1) - c S(I_5) + \frac{c \nu_p}{2} I_5 + \frac{\bar{n} - \bar{\nu}}{2} T_4^H - \frac{\bar{n}}{2} T_5^H & \text{if } J = 1, \\ +\infty & \text{otherwise}. \end{cases} \quad (10.16)$$

with

$$I_1 = \bar{s}[I_1 - N] + N \quad \text{and} \quad I_5 = -\frac{1}{c} \left[ \frac{\partial \bar{m}}{\partial \nu_p} - \frac{\partial \bar{H}}{\partial \nu_p} \right] T_4^H + \frac{1}{c} \frac{\partial \bar{m}}{\partial \nu_p} T_5^H. \quad (10.17)$$

\(^4\)The shear modulus of iron is in the order of hundreds of GPa, whereas the shear modulus of a conventional rubber is, at most, in the order of MPa. On the other hand, ferrofluids are colloidal suspensions of ferromagnetic nanoparticles in a carrier fluid that exhibit near incompressibility and close-to-zero resistance to shear.
Here,

\[
\begin{align*}
\tilde{s} &= \frac{2}{(N^2 + N - 2)(1 - c)} G \int_{\Omega} g(X) K_{klmn} \Gamma_{mkl,n} dX, \\
\tilde{\nu} &= \frac{1}{N} \int_{\Omega} \mu(X) \gamma_{m,m} dX, \\
\tilde{n} &= \frac{2}{N^2 + N - 2} \int_{\Omega} \mu(X) K_{ijkl} \Gamma_{rij,s} K_{rsuv} \gamma_{u,k} \gamma_{v,l} dX,
\end{align*}
\]

(10.18)

with \( g(X) = [1 - \theta(X)] G + \theta(X) G_\text{p} \) and \( \mu(X) = [1 - \theta(X)] \mu_0 + \theta(X) \nu_p \), where the coefficient \( \nu_p \geq \mu_0 \) is defined implicitly as solution of the nonlinear algebraic equation

\[
2S'(I_5) - \nu_p = 0,
\]

(10.19)

\( K_{ijkl} = \frac{1}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) - \frac{1}{N} \delta_{ij} \delta_{kl}, \quad \delta_{ij} \) denoting the Kronecker delta, and the tensor fields \( \Gamma(X) \) and \( \gamma(X) \) are defined as the solutions of the uncoupled linear boundary value problems

\[
\begin{align*}
&[[g(X) K_{ijkl} \Gamma_{mkl,n} + \delta_{ij} q_{kl}]]_j = 0, & X \in \Omega \\
&\Gamma_{mkl,m} = 0, & X \in \Omega \\
&\Gamma_{ikl} = \delta_{ik} X_l, & X \in \partial \Omega
\end{align*}
\]

(10.20)

and

\[
\begin{align*}
&[[\mu(X) \gamma_{i,j}]]_i = 0, & X \in \Omega \\
&\gamma_i = X_i, & X \in \partial \Omega
\end{align*}
\]

(10.21)

In the above expressions, the notation \( _i \) represents partial differentiation with respect to the material point coordinate \( X_i \), \( q(X) \) is a tensorial field associated with the incompressibility constraint \( \Gamma_{mkl,m} = 0 \) in \( \Omega \), and we recall again that \( N \) stands for the space dimension \( (N = 2, 3) \), while \( I_1, T_4^H, T_5^H \) stand for the macroscopic invariants defined by expressions (10.15)\textsubscript{1,3,4}. We refer the interested reader to Section 8.1 in Chapter 8 for the derivation (for \( N = 3 \)) of the variational solution (10.16) as well as for a detailed description of its features. Here, it suffices to record the following remarks:

\[ \text{i. The macroscopic constitutive magnetomechanical relation (10.10) implied by the free-energy function (10.16) is given by} \]

\[
\mathbf{S} = 2(1 - c)\tilde{s} \Psi' I_1 \mathbf{F} + \tilde{n} \mathbf{F}^{-T} \mathbf{H} \otimes \mathbf{F}^{-1} \mathbf{F}^{-T} \mathbf{H} - \rho \mathbf{F}^{-T},
\]

(10.22)
where $p$ stands for the arbitrary hydrostatic pressure associated with the macroscopic incompressibility constraint $J = 1$, and

$$
\mathbf{B} = (\bar{\nu} - \bar{n}) \mathbf{H} + \bar{n} \mathbf{F}^{-1} \mathbf{F}^{-T} \mathbf{H}.
$$

(10.23)

In turn, the macroscopic Cauchy stress, macroscopic Eulerian magnetic induction, and macroscopic magnetization are given by

$$
\mathbf{T} = 2(1 - c)\bar{s} \Psi'(I_1) \mathbf{F} \mathbf{F}^T + \bar{n} \mathbf{F}^{-T} \mathbf{H} \otimes \mathbf{F}^{-T} \mathbf{H} - p \mathbf{I},
$$

$$
\mathbf{b} = (\bar{\nu} - \bar{n}) \mathbf{F} \mathbf{H} + \bar{n} \mathbf{F}^{-T} \mathbf{H},
$$

$$
\mathbf{m} = \frac{\bar{\nu} - \bar{n}}{\mu_0} \mathbf{F} \mathbf{H} + \frac{\bar{n} - \mu_0}{\mu_0} \mathbf{F}^{-T} \mathbf{H},
$$

(10.24)

respectively.

ii. Evaluation of the formula (10.16) for the effective free energy $W$ and of the formulas (10.22) and (10.23) for the macroscopic constitutive relations requires knowledge of the coefficients $\bar{s}$, $\bar{\nu}$, $\bar{n}$, $\nu_p$. All four of them depend on the constitutive behaviors of the rubber and particles through the material functions/parameters $\Psi$, $\mu_0$, $G_p$, $S$ and on the microstructure through the solutions $\Gamma(X)$ and $\gamma(X)$ of the pdes (10.20)–(10.21). In addition, the coefficients $\bar{\nu}$, $\bar{n}$, $\nu_p$ depend as well on the magnetomechanical loading through the invariants $I_4^H$ and $I_5^H$.

For a given choice of material functions/parameters $\Psi$, $\mu_0$, $G_p$, $S$ and a given isotropic microstructure, the coefficients $\bar{s}$, $\bar{\nu}$, $\bar{n}$, $\nu_p$ can be obtained as follows. First, the pde (10.20) is solved for $\Gamma(X)$. In general, this pde as well as the pde (10.21) for the field $\gamma(X)$ do not admit analytical solutions, but can be readily solved numerically using finite elements; see Sections 5.2 and 5.1 for details in $N = 2$ and 3 space dimensions. Knowledge of $\Gamma(X)$ then allows for the evaluation by means of a quadrature rule of the integral (10.18)$_1$ that defines the effective coefficient $\bar{s}$. As a second step, the pde (10.21) is solved for $\gamma(X)$ multiple times for a sufficiently wide range of values of $\nu_p \geq \mu_0$ so as to allow for the numerical computation of the derivatives $\partial \bar{\nu}/\partial \nu_p$ and $\partial \bar{n}/\partial \nu_p$ entering in (10.17)$_2$, the numerical solution of the nonlinear algebraic equation (10.19) defining $\nu_p$, and the evaluation by means of a quadrature rule of the integrals (10.18)$_{2,3}$ defining the effective coefficients $\bar{\nu}$ and $\bar{n}$.

In practice, from the above-described numerical construction, it is possible to obtain explicit interpolating formulas for the coefficients $\bar{\nu}$ and $\bar{n}$ in terms of the coefficient $\nu_p$. Having access to these formulas reduces the computation of the effective energy (10.16) and corresponding
constitutive relations (10.22) and (10.23) simply to solving the nonlinear algebraic equation (10.19) for $\nu_p$. We report such explicit formulas for the basic cases of isotropic suspensions of circular and spherical particles in Sections 10.3 and 10.4.

iii. By construction, the variational solution (10.16) is asymptotically exact in the limit of small deformations and moderate magnetic fields, namely, when $\varepsilon_{ij} = \mathbf{F}_{ij} - \delta_{ij} = O(\zeta)$ and $\vec{H}_{ij} = O(\zeta^{1/2})$ for a vanishingly small parameter $\zeta$ (Tian et al., 2012; Chapter 7). In this limit, the nonlinear algebraic equation (10.19) admits the explicit solution $\nu_p = 2S'(0) = \mu_p$ to leading order, and the effective free energy (10.16) reduces asymptotically to

$$W(\mathbf{F}, \vec{H}) = \begin{cases} (1 - c)G \bar{\nu} \bar{\varepsilon} \cdot \bar{\varepsilon} - \nabla \mu_p \vec{H} \cdot \vec{H} + \bar{n} \vec{H} \cdot \bar{\varepsilon} \bar{H} & \text{if } \text{tr} \bar{\varepsilon} = 0 \\ +\infty & \text{otherwise} \end{cases}$$

(10.25)

where, again, the effective coefficients $\bar{\nu}$, $\bar{\varepsilon}$, $\bar{n}$ are given by relations (10.18) with $\nu_p = \mu_p$.

While, in general, the variational solution (10.16) is not exact for finite deformations and finite magnetic fields, direct comparisons with full-field simulations for the case of $N = 3$ have shown that it remains accurate for arbitrary magnetomechanical loadings (see Section 8.4 in Chapter 8). The accuracy of the variational solution (10.11) for finite deformations and finite magnetic fields for the case of $N = 2$ space dimensions is demonstrated below in Section 10.3 by analogous comparisons with full-field simulations.

iv. For the fundamental limiting case when the underlying filler particles are made of a linear magnetic material, so that

$$S(t^H_5) = \frac{\mu_p}{2} t^H_5,$$

(10.26)

the equation (10.19) admits the explicit solution $\nu_p = \mu_p$ and the effective free-energy function (10.16) reduces rather simply to

$$W(\mathbf{F}, \vec{H}) = \begin{cases} (1 - c)\Psi(\mathcal{I}_1) + \frac{\bar{n} - \bar{\nu}}{2} \mathcal{T}^H_4 - \frac{\bar{n}}{2} \mathcal{T}^H_5 & \text{if } \mathcal{J} = 1 \\ +\infty & \text{otherwise} \end{cases}$$

(10.27)

where, again, $\mathcal{I}_1$ is given by expression (10.17) and the effective constants $\bar{\nu}$ and $\bar{n}$ are given by relations (10.18) with $\nu_p = \mu_p$.

v. Depending on the specific problem at hand, it might be more convenient to employ the macroscopic Lagrangian magnetic induction $\mathbf{B}$ as the independent macroscopic magnetic variable,
instead of the magnetic field $\mathbf{H}$. Given that the concavity of the free-energy functions (10.1) and (10.2) in $\mathbf{H}$ implies that the effective free-energy function (10.16) is concave in $\mathbf{H}$ within a possibly unbounded neighborhood of $\mathbf{F} = \mathbf{I}$, this can be readily accomplished with help of the partial Legendre transform

$$W^*(\mathbf{F}, \mathbf{B}) = \sup_{\mathbf{H}} \left\{ \mathbf{B} \cdot \mathbf{H} + W(\mathbf{F}, \mathbf{H}) \right\},$$

(10.28)

from which the macroscopic Piola-Kirchhoff stress and the macroscopic Lagrangian magnetic field can be written in terms of $\mathbf{B}$ as follows:

$$\mathbf{S} = \frac{\partial W^*(\mathbf{F}, \mathbf{B})}{\partial \mathbf{F}}(\mathbf{F}, \mathbf{B}) \quad \text{and} \quad \mathbf{H} = \frac{\partial W^*(\mathbf{F}, \mathbf{B})}{\partial \mathbf{B}}(\mathbf{F}, \mathbf{B}).$$

(10.29)

Physically, the free energy (10.28) corresponds to the effective Helmholtz free energy of the magnetorheological elastomer.

As an illustrative example, the partial Legendre transform (10.28) of the effective free energy (10.27) for magnetorheological elastomers with linear magnetic particles renders the effective Helmholtz free energy

$$W^*(\mathbf{F}, \mathbf{B}) = \begin{cases} (1 - c)\Psi(I_1) + \frac{1}{2\tilde{n}} \left[ \tilde{\eta}I_4^B + \tilde{I}_5^B \right] & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases}$$

(10.30)

for $N = 2$, and

$$W^*(\mathbf{F}, \mathbf{B}) = \begin{cases} (1 - c)\Psi(I_1) + \frac{1}{2\tilde{n}} \left[ \tilde{\eta}I_4^B + \tilde{\eta}I_1I_5^B - I_6^B \right] & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases}$$

(10.31)

for $N = 3$. In these last expressions, the coefficient $\tilde{\eta} = (\tilde{\nu} - \tilde{n})/\tilde{n}$ has been introduced to ease notation, $I_4^B$, $I_5^B$, $I_6^B$ stand for the standard invariants

$$I_4^B = \mathbf{B} \cdot \mathbf{B}, \quad I_5^B = \mathbf{F}^B \cdot \mathbf{F}^B, \quad I_6^B = \mathbf{F}^T \mathbf{F}^B \cdot \mathbf{F}^T \mathbf{F}^B,$$

(10.32)

and it is recalled that $I_1$ is given by expression (10.17)$_1$, while the effective constants $\tilde{\nu}$ and $\tilde{n}$ are given by (10.18)$_{2,3}$ with $\nu_p = \mu_p$. 
vi. We remark that for the case of \( N = 2 \) space dimensions, the (finite branch of the) effective free energy (10.16) is of the \textit{separable} form \( W = W_{\text{elas}}(I_1) + W_{\text{mag}}(I_4^H, I_5^H) \). By contrast, when written in terms of \( \mathbf{F} \) and \( \mathbf{B} \) as the independent variables, it is not difficult to deduce that the (finite branch of the) corresponding Helmholtz free energy (10.28) is of the general \textit{non-separable} form \( W^* = W^*(I_1, I_4^B, I_5^B) \); see, for instance, the Helmholtz free energy (10.30).

For the case of \( N = 3 \), the (finite branch of the) effective free energy (10.16) is also of the \textit{separable} form \( W = W_{\text{elas}}(I_1) + W_{\text{mag}}(I_4^H, I_5^H) \), but, interestingly, it only depends on three of the five isotropic invariants (10.15). By contrast, the (finite branch of the) corresponding Helmholtz free energy (10.28) is of the general \textit{non-separable} form \( W^* = W^*(I_1, I_2, I_4^B, I_5^B, I_6^B) \); see, for instance, the Helmholtz free energy (10.31).

The above-outlined homogenization-based functional dependencies on the standard invariants \( I_1, I_2, I_4^H, I_5^H, I_6^H, I_4^B, I_5^B, I_6^B \) differ from the phenomenological ones that have been suggested/utilized in the literature based on the limited available experimental data; see, e.g., the works of Dorfmann and Ogden, (2005b), Bustamante et al., (2011), and Pelteret et al., (2016).

### 10.3 The basic case of an isotropic suspension of \textit{circular} particles

In this section, we present the specialization of the effective free energy (10.16) to the basic case in \( N = 2 \) space dimensions of magnetorheological elastomers wherein the isotropically distributed filler particles are monodisperse in size and \textit{circular} in shape. We begin by presenting in subsection 10.3.1 the result for circular iron particles and confront it with full-field simulations to demonstrate its accuracy for finite deformations and finite magnetic fields. The same is done in subsection 10.3.2 for the specialization to circular ferrofluid particles.

Before proceeding with the presentation of the results, we recall that explicit interpolating formulas for the coefficients \( \tilde{\nu} \) and \( \tilde{n} \) in terms of the coefficient \( \nu_p \) can be obtained via the numerical construction outlined in remark \( \text{ii} \) of Section 10.2. Repeating these calculations for various volume fractions of particles \( c \) allows, moreover, to obtain explicit interpolating formulas for \( \tilde{\nu} \) and \( \tilde{n} \), as well as for the coefficient \( \tilde{s} \), in terms of \( c \). We shall present below such formulas for iron as well as for ferrofluid circular particles. Again, having access to these formulas reduces the computation of the
effective energy (10.16), and the corresponding constitutive relations (10.22) and (10.23), simply to solving the nonlinear algebraic equation (10.19) for \( \nu_p \).

### 10.3.1 The solution for circular iron particles

Figure 10.2 shows plots for the case of circular iron particles \( (G_p = +\infty) \) of the coefficient \( \tilde{s} \) defined by expression (10.18) as a function of the volume fraction of particles \( c \), as well as of the coefficients \( \tilde{\nu} \) and \( \tilde{n} \) defined by expression (10.18) as functions of \( c \) and the coefficient \( \nu_p \). The solid circles (labeled as "Rig. Cir. FE" in the plots) correspond to results based on finite-element solutions of the pdes (10.20) and (10.21) for the fields \( \Gamma(X) \) and \( \gamma(X) \), while the solid line and solid surfaces (labeled as "Rig. Cir. Analytical" in the plots) correspond to the following explicit interpolating formulas:

\[
\begin{align*}
\tilde{s} &= (1 - c)^{-3}, \\
\tilde{\nu} &= \mu_0 + \frac{2c\mu_0(\nu_p - \mu_0)}{(1 + c)\mu_0 + (1 - c)\nu_p}, \\
\tilde{n} &= \mu_0 + \frac{c(8 + 4c + 3c^2 + c^3)(\nu_p - \mu_0)\mu_0^2}{4[(1 - c)\nu_p + (1 + c)\mu_0]^2} + \frac{c(1 - c)(8 + 4c + c^2)(\nu_p - \mu_0)\mu_0\nu_p}{4[(1 - c)\nu_p + (1 + c)\mu_0]^2}.
\end{align*}
\]

(10.33)

By construction, these formulas are valid for all values of \( \nu_p \geq \mu_0 \) and the range of volume fractions \( c \in [0, 0.35] \). Their functional forms are inspired from existing analytical solutions for differential coated cylinder assemblages (see Section 4.2) and have the merit that they are asymptotically exact in the dilute limit of particles as \( c \to 0^+ \); note that in this limit, the linear pdes (10.20) and (10.21) do admit an analytical solution.

Making direct use of the formulas (10.33) for the coefficients \( \tilde{s}, \tilde{\nu}, \tilde{n} \) for circular iron particles in the general result (10.16) renders the following effective free-energy function:

\[
W(F, H) = \begin{cases} 
(1 - c)\Psi\left(\tilde{I}_1^{\text{Cir}}\right) - cS\left(\tilde{I}_5^{\text{Cir}}\right) + \frac{c\nu_p}{2}\tilde{I}_5^{\text{Cir}} + \frac{\tilde{n} - \tilde{\nu}}{2}I_4^H - \frac{\tilde{n}}{2}I_5^H & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]

(10.34)

with

\[
\begin{align*}
\tilde{I}_1^{\text{Cir}} &= \frac{\tilde{I}_1 - 2}{(1 - c)^3} + 2, \\
\tilde{I}_5^{\text{Cir}} &= \frac{c(1 - c)(4 + c)(\nu_p - \mu_0)\mu_0^2}{[(1 + c)\mu_0 + (1 - c)\nu_p]^3}I_4^H + \frac{[(4 + 3c^2 + c^3)\mu_0 + (1 - c)(2 + c)^2\nu_p]\mu_0^2}{[(1 + c)\mu_0 + (1 - c)\nu_p]^3}I_5^H.
\end{align*}
\]

(10.35)

(10.36)

\(^6\)The finite-element results presented in Fig. 10.2, and in Fig. 10.4 below, correspond to the average of three different realizations of a square unit cell, repeated periodically ad infinitum, that contains a random distribution of 60 circular particles. In the context of the pdes (10.20) and (10.21), realizations with 60 circular particles per unit cell provide an accurate approximation of a truly random isotropic distribution of circular particles (see, e.g., Section 5.2).
Figure 10.2: Plots of the coefficients \( \tilde{s} \), \( \tilde{\nu} \), \( \tilde{n} \) defined by expressions (10.18) for the case of circular iron particles \( (G_p = +\infty) \). Part (a) shows the coefficient \( \tilde{s} \) as a function of the volume fraction of particles \( c \). Parts (b) and (c) show the normalized coefficients \( \tilde{\nu}/\mu_0 \) and \( \tilde{n}/\mu_0 \) as functions of \( c \) and the normalized coefficient \( \mu_0/\nu_p \). The solid circles (“Rig. Cir. FE”) correspond to finite-element results, while the solid line and solid surfaces (“Rig. Cir. Analytical”) correspond to the explicit interpolating formulas (10.33).

where the coefficient \( \nu_p \) is now defined implicitly as solution of the nonlinear algebraic equation

\[
2S' \left( I_{Cir} \right) - \nu_p = 0. \tag{10.37}
\]

Here, we re-emphasize that the free energy (10.34) is fully explicit up to the above nonlinear algebraic equation for \( \nu_p \), which requires, in general, a numerical treatment.

### 10.3.1.1 Comparisons with full-field simulations

Next, we illustrate the accuracy of the effective free-energy function (10.34) for finite deformations and finite magnetic fields by sample comparisons with full-field simulations. For definiteness, we take the function \( \Psi \) in the free energy (10.1) characterizing the constitutive behavior of the rubber matrix to be given by (10.5) with the physically realistic parameters \( G_1 = 0.1 \text{ MPa}, G_2 = 0 \text{ MPa}, \alpha_1 = \alpha_2 = 1 \) (Lopez-Pamies, 2010). Moreover, we take the volume fraction of iron particles at
$c = 0.15$ and the function $S$ in the free energy (10.2) characterizing their constitutive behavior to be given by (10.8) with the physically realistic parameters $\mu_p = 100\mu_0$ and $m_s = 1$ MA/m ((Arias et al., 2006). The full-field simulations reported here are entirely analogous to those put forth in Chapter 8 for $N = 3$ space dimensions. Namely, they are generated by means of a conforming 7-node hybrid triangular finite element discretization of the Euler-Lagrange equations (10.12) for an infinite medium made up of the periodic repetition of a square unit cell containing a large but finite random distribution of circular particles. All the results based on full-field simulations that are presented here, and below in subsection 10.3.2.1, correspond to the average of three realizations with 60 particles per unit cell. In the context of the pdes (10.12), realizations with 60 particles per unit cell have been checked to provide an adequate approximation of a truly random isotropic distribution of circular particles (at least up to the maximum value of volume fractions of particles considered here $c = 0.35$).

Figure 10.3: Plots of the effective free-energy function $W$ for a rubber isotropically filled with a volume fraction $c = 0.15$ of circular iron particles. The results are shown in terms of each one of the invariants $I_1$, $I_H^4$, $I_H^5$ for two sets of fixed values of the remaining two invariants. The solid lines (“Rig. Cir. Theory”) correspond to the free-energy function (10.34), while the dashed lines (“Rig. Cir. FE”) correspond to the full-field simulations.

Figure 10.3 displays the comparisons between the effective free-energy function (10.34) and corresponding full-field simulations. To aid in the visualization of their quantitative agreement for finite deformations and finite magnetic fields, the results are shown as a function of each one of the invariants $I_1$, $I_H^4$, $I_H^5$, while the remaining two invariants are kept fixed. In this regard, we notice that fixing the values of $I_H^4$ and $I_H^5$ bounds from below the range of physical values that $I_1$ can take on. On the other hand, fixing the values of $I_1$ and either $I_H^4$ or $I_H^5$ bounds from below and from above the range of values that the remaining magnetic invariant can physically take on; for example,
\( T_1 \geq 2.02 \) for the fixed values \( T_4^H = 0.32 \text{ MA}^2/\text{m}^2 \) and \( T_5^H = 0.37 \text{ MA}^2/\text{m}^2 \), while \( T_4^H \in [0.22, 0.62] \) MA\(^2/\text{m}^2 \) for the fixed values \( T_1 = 2.28 \) and \( T_5^H = 0.37 \text{ MA}^2/\text{m}^2 \). The results displayed in Fig. 10.3(a) correspond to physically allowable values of \( T_1 \) from its lower bound up to the point at which we managed to have convergence in our full-field simulations. On the other hand, the results shown in Figs. 10.3(b) and (c) span the entire range of allowable values for \( T_4^H \) and \( T_5^H \). We further remark that the selected values of the macroscopic invariants \( T_1, T_4^H, T_5^H \) are representative of conventional experimental capabilities and involve values of the local invariant \( T_5^H(X) \) that are large enough in the iron particles so as to trigger the saturation of their magnetization.

A quick glance at the plots in Fig. 10.3 suffices to recognize that the free energy (10.34) is in good agreement with the full-field simulations for the selected values of the constitutive, geometric, and loading parameters. A large body of results, not reported here for conciseness, has confirmed that the free energy (10.34) remains in agreement with full-field simulations for arbitrary magnetomechanical loadings, irrespectively of the constitutive properties of the rubber matrix and iron particles.

### 10.3.2 The solution for circular ferrofluid particles

For the case of circular ferrofluid particles \((G_p = 0)\), the coefficients \( \tilde{s}, \tilde{\nu}, \tilde{n} \) defined by expressions (10.18) can be accurately approximated, for all values \( \nu_p \geq \mu_0 \) and the range of volume fraction of particles \( c \in [0, 0.35] \), by the formulas

\[
\tilde{s} = (1 - c), \quad \tilde{\nu} = \mu_0 + \frac{2c\mu_0(\nu_p - \mu_0)}{(1 + c)\mu_0 + (1 - c)\nu_p}, \\
\tilde{n} = \mu_0 + \frac{4c^2(\nu_p - \mu_0)^2\mu_0}{[(1 - c)\nu_p + (1 + c)\mu_0]^2} \left[ \frac{\mu_0}{c(\nu_p - \mu_0)} + (1 - c) \left( \frac{1}{4} + \frac{133}{500} c^{1/4} - \frac{213}{250} c^{1/2} \right) \right].
\]  

(10.38)

Akin to the explicit coefficients (10.33), the explicit coefficients (10.38) correspond to interpolating formulas for a set of numerical values of expressions (10.18) based on finite-element solutions of the pdes (10.20) and (10.21) for the fields \( \Gamma(X) \) and \( \gamma(X) \). Furthermore, the functional forms of the explicit coefficients (10.38) are also inspired from existing analytical solutions for differential coated cylinder assemblages and have the merit that they are asymptotically exact in the dilute limit of particles as \( c \to 0^+ \). Figure 10.4 shows plots, in terms of \( c \) and \( \nu_p \), of the formulas (10.38) for the coefficients \( \tilde{s}, \tilde{\nu}, \tilde{n} \) and the corresponding numerical results based on the finite-element solutions for \( \Gamma(X) \) and \( \gamma(X) \).

Making direct use of the formulas (10.38) for the coefficients \( \tilde{s}, \tilde{\nu}, \tilde{n} \) for circular ferrofluid particles
in the general result (10.16) yields the following effective free-energy function:

\[
W(F, H) = \begin{cases} 
(1 - c)\Psi \left( T_1^{\text{Cir}} \right) - c S \left( T_5^{\text{Cir}} \right) + \frac{c \nu_p T_5^{\text{Cir}}}{2} - \frac{\tilde{n}}{\nu_0} \tilde{T}_4 - \frac{\tilde{n}}{2} \tilde{T}_5 & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]  

(10.39)

with

\[
T_1^{\text{Cir}} = (1 - c)[T_1 - 2] + 2,  
T_5^{\text{Cir}} = \frac{16(1 - c)(\nu_p - \mu_0)\mu_0^2}{(1 + c)\mu_0 + (1 - c)\nu_p} \left[ \frac{(1 + c)\mu_0 + (1 - c)\nu_p}{4(1 - c)(\nu_p - \mu_0)} \tilde{T}_5 + \left( \frac{1}{2} - 1 \frac{133}{200} c^{5/4} + \frac{213}{250} c^{3/2} \right) \left( \tilde{T}_4 - \tilde{T}_5 \right) \right].
\]  

(10.41)
where the coefficient $\nu_p$ is defined implicitly by the nonlinear algebraic equation

$$2\mathcal{S}'\left(T_5^{\text{Cir}}\right) - \nu_p = 0.$$  \hspace{1cm} (10.42)

Here too we note that the free energy (10.39) is fully explicit up to the nonlinear algebraic equation (10.42) for $\nu_p$.

### 10.3.2.1 Comparisons with full-field simulations

The plots presented in Fig. 10.5 provide sample comparisons between the effective free energy (10.39) and corresponding full-field simulations. The results correspond to the same rubber matrix and the same volume fraction of particles as those utilized in Fig. 10.3 for the case of circular iron particles, but in this case the function $\mathcal{S}$ in the free energy (10.2) characterizing the constitutive behavior of the particles is given by (10.8) with the material parameters $\mu_p = 10\mu_0$ and $m_s = 0.05$ MA/m, which are representative of standard ferrofluids (Kuzhir et al., 2006; Ivanov et al., 2007).

In particular, Fig. 10.5(a) shows the effective free energy as a function of $T_1$ for two sets of fixed values of $T_4^H$ and $T_5^H$, whereas Figs. 10.5(b) and (c) show the effective free energy as a function of $T_4^H$ and $T_5^H$ for two sets of fixed values of the pairs $T_1$, $T_5^H$ and $T_1$, $T_4^H$. We note that these sets of results correspond to physically allowable values of the invariants (recall that fixing two out of the three invariants $T_1, T_4^H, T_5^H$ bounds the range of values that the remaining invariant can take on) that involve values of the local invariant $I_5^H(X)$ that are large enough in the ferrofluid particles so as to trigger the saturation of their magnetization.

It is plain from Fig. 10.5 that the free energy (10.39) is in good quantitative and qualitative agreement with the full-field simulations for the selected values of the constitutive, geometric, and loading parameters. A spectrum of results beyond those reported here has confirmed that the free energy (10.39) remains in agreement with full-field simulations for arbitrary magnetomechanical loadings, irrespectively of the constitutive properties of the rubber matrix and ferrofluid particles.

### 10.4 The basic case of an isotropic suspension of spherical particles

In the sequel, we report the specialization of the free energy (10.16) to the basic case in $N = 3$ space dimensions of magnetorheological elastomers wherein the isotropically distributed filler particles
10. Magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber

Figure 10.5: Plots of the effective free-energy function $W$ for a rubber isotropically filled with a volume fraction $c = 0.15$ of circular ferrofluid particles. The results are shown in terms of each one of the invariants $T_1, T_4^H, T_5^H$ for two sets of fixed values of the remaining two invariants. The solid lines (“Liq. Cir. Theory”) correspond to the free-energy function (10.39), while the dashed lines (“Liq. Cir. FE”) correspond to the full-field simulations.

are monodisperse in size and **spherical** in shape. We present the result for spherical iron particles in subsection 10.4.1 and that for spherical ferrofluid particles in subsection 10.4.2. We note that the accuracy of both of these results has already been demonstrated — within the mathematically analogous setting of elastic dielectric composites — for finite deformations and finite magnetic fields via direct comparisons with full-field simulations in Section 8.4. Accordingly, we do not reproduce such comparisons here.

### 10.4.1 The solution for spherical iron particles

For the case of spherical iron particles ($G_p = +\infty$), the coefficients $\tilde{s}, \tilde{\nu}, \tilde{n}$ defined by expressions (10.18) can be accurately approximated, for all values of the coefficient $\nu_p \geq \mu_0$ and the range of volume fraction of particles $c \in [0, 0.25]$, by the formulas

$$\tilde{s} = (1 - c)^{-7/2}, \quad \tilde{\nu} = \mu_0 + \frac{3c\mu_0(\nu_p - \mu_0)}{(2 + c)\mu_0 + (1 - c)\nu_p},$$

$$\tilde{n} = \mu_0 + \frac{3c(10 + 2c + 3c^2)(\nu_p - \mu_0)\mu_0^2}{5[(2 + c)\mu_0 + (1 - c)\nu_p]^2} + \frac{3c(1-c)(5+3c)(\nu_p - \mu_0)\mu_0\nu_p}{5[(2 + c)\mu_0 + (1 - c)\nu_p]^2}. \quad (10.43)$$

Upon direct use of these explicit expressions, the general result (10.16) specializes to the following effective free-energy function:

$$W(F,H) = \begin{cases} 
(1-c)\Psi \left( I_1^{\text{Sph}} \right) - cS \left( I_5^{\text{Sph}} \right) + \frac{c\nu_p}{2} I_5^{\text{Sph}} + \frac{\tilde{n} - \tilde{\nu}}{2} T_4^H - \frac{\tilde{n}}{2} T_5^H & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases} \quad (10.44)$$
10 Magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber

\[
I_{\text{Sph}r} = \frac{I_1 - 3}{(1 - c)^{7/2}} + 3, \quad (10.45)
\]

\[
I_{\text{Sph}r}^5 = -\frac{54c(1 - c)\mu_0^2}{5[(2 + c)\mu_0 + (1 - c)\mu_p]^3} + \frac{9[(10 - c + 6c^2)\mu_0 + (5 + c - 6c^2)\nu_p]\mu_0^2}{5[(2 + c)\mu_0 + (1 - c)\mu_p]^3} + \frac{9}{5} \left( \frac{2 + c}{\mu_0} + \frac{1 - c}{\nu_p} \right), \quad (10.46)
\]

where the coefficient \(\nu_p\) is defined implicitly as solution of the nonlinear algebraic equation

\[
S' \left( I_{\text{Sph}r}^5 \right) - \frac{\nu_p^2}{2} = 0. \quad (10.47)
\]

Similar to its counterpart (10.34) for circular iron particles, the free energy (10.44) is fully explicit up the nonlinear algebraic equation (10.47) for \(\nu_p\).

10.4.2 The solution for spherical ferrofluid particles

Finally, for the case of spherical ferrofluid particles \((G_p = 0)\), the coefficients \(\tilde{s}, \tilde{\nu}, \tilde{n}\) defined by expressions (10.18) can be accurately approximated by the formulas

\[
\tilde{s} = (1 - c)^{2/3}, \quad \tilde{\nu} = \mu_0 + \frac{3c\mu_0(\nu_p - \mu_0)}{(2 + c)\mu_0 + (1 - c)\nu_p},
\]

\[
\tilde{n} = \mu_0 + \frac{9c^2(\nu_p - \mu_0)^2\mu_0}{[(2 + c)\mu_0 + (1 - c)\nu_p]^2} \left( \frac{4}{45} - \frac{81^{1/25}}{500} + \frac{\mu_0}{c(\nu_p - \mu_0)} \right), \quad (10.48)
\]

which are valid for all values of the coefficient \(\nu_p \geq \mu_0\) and the range of volume fraction of particles \(c \in [0, 0.25]\). Much like the explicit coefficients (10.43), the explicit coefficients (10.48) correspond to interpolating formulas based on a set of numerical values of expressions (10.18) generated from finite-element solutions of the pdes (10.20) and (10.21) for the fields \(\Gamma(X)\) and \(\gamma(X)\). The coefficients (10.48) also draw their functional forms from existing analytical solutions for differential coated sphere assemblages (see Section 4.1) and are asymptotically exact in the dilute limit as \(c \to 0^+\).

Direct use of the formulas (10.48) for the coefficients \(\tilde{s}, \tilde{\nu}, \tilde{n}\) for spherical ferrofluid particles in the general result (10.16) leads to the following effective free-energy function:

\[
\mathcal{W}(\mathbf{F}, \mathbf{H}) = \begin{cases} 
(1 - c)\Psi \left( I_{\text{Sph}r}^1 \right) - c\mathcal{S} \left( I_{\text{Sph}r}^5 \right) + \frac{c\nu_p^2}{2} I_{\text{Sph}r}^5 + \frac{\tilde{n} - \tilde{\nu}}{2} I_4^H - \frac{\tilde{n}}{2} I_5^H & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]

(10.49)
with

$$I_{1}^{Sph} = (1 - c)^{2/3}(I_{1} - 3) + 3, \quad (10.50)$$

$$I_{5}^{Sph} = \frac{3(1500 - 1900c + 729c^{36/25})(\nu_{p} - \mu_{0})\mu_{0}^{2}}{250[(2 + c)\mu_{0} + (1 - c)\nu_{p}]^{3}}I_{4}^{H} - \frac{3[(750 - 1150c + 729c^{36/25})(\nu_{p} - \mu_{0}) - 2250\mu_{0}]\mu_{0}^{2}}{250[(2 + c)\mu_{0} + (1 - c)\nu_{p}]^{3}}I_{5}^{H}, \quad (10.51)$$

where the coefficient $\nu_{p}$ is now defined implicitly by the nonlinear algebraic equation

$$S'(I_{5}^{Sph}) - \frac{\nu_{p}}{2} = 0. \quad (10.52)$$

Similar to its counterpart (10.39) for circular ferrofluid particles, the effective free energy (10.49) is fully explicit up to the numerical treatment required, in general, to compute the coefficient $\nu_{p}$ from its implicit definition (10.52).

### 10.5 Iron particles vs. ferrofluid particles

Since the classical work of Brown (1966), it is well known that a homogeneous magnetoelastic specimen of ellipsoidal shape does not deform uniformly when exposed to a remotely applied homogeneous magnetic field. In spite of the fundamental nature of this Eshelby-type boundary-value problem, its solution does not appear to have been reported in the literature; see, however, Sections 10.2–10.4 of Chapter IV in Brown (1966) and references therein for a number of partial solutions in the asymptotic context of small deformations. Leveraging the general result (10.16) for the effective free energy of isotropic magnetorheological elastomers, the compound objective of this section is to present such a solution — which, by necessity, is numerical — and to gain insight into the magnetoelastic behavior of magnetorheological elastomers containing ferrofluid filler particles vis-à-vis that of magnetorheological elastomers containing iron filler particles.

For definiteness, we consider the boundary-value problem of a specimen of initial spherical shape and unit radius immersed in an infinite medium otherwise filled by air that is subjected to a remotely applied homogeneous magnetic field, $H_{\infty}$ say. The specimen is made up of an isotropic magnetorheological elastomer whose magnetoelastic behavior is characterized by either the effective free-energy function (10.44) corresponding to spherical iron filler particles or the effective free-energy function

---

7Specifically, the classical result of Brown (1966) applies to finite deformations, finite magnetic fields, and any anisotropic magnetoelastic solid featuring orthotropic material symmetry; see Section 10.1 of Chapter IV in Brown (1966).
Figure 10.6: (a) Schematic of the finite domain utilized to generate numerical solutions for the boundary-value problem (10.54). The air domain is defined by an initial outer radius that is twenty times that of the spherical specimen. (b) Detail of the corresponding axisymmetric discretization with 7-node hybrid triangular finite elements.

(10.49) corresponding to spherical ferrofluid filler particles. For convenience, among the various possible modeling strategies, we opt to model the surrounding air as a compressible magnetoelastic material of vanishing small mechanical stiffness with free-energy function

\[ W_a(F, H) = \frac{G_a}{2} \left[ I_1 - 3 - 2 \ln J \right] - \frac{\mu_0}{2} J I_5^H \] with \[ G_a = 0^+ , \] (10.53)

where we recall that \[ I_1 = F \cdot F, \quad J = \det F, \quad I_5^H = F^{-T} H \cdot F^{-T} H, \] and \[ \mu_0 = 4\pi \times 10^{-7} \text{ H/m} . \] Thus, more precisely, we are interested in solving the boundary-value problem

\[
\begin{align*}
\text{Div } S(X) &= 0, \quad X \in \mathbb{R}^3 \\
\text{Div } B(X) &= 0, \quad X \in \mathbb{R}^3 \\
\chi(X) &= X, \quad ||X|| \to \infty \\
\psi(X) &= -H_{\infty} \cdot X, \quad ||X|| \to \infty
\end{align*}
\] (10.54)

where

\[ S(X) = \theta(X) \frac{\partial W}{\partial F}(F, H) + (1 - \theta(X)) \frac{\partial W_a}{\partial F}(F, H) \] (10.55)

and

\[ B(X) = -\theta(X) \frac{\partial W}{\partial H}(F, H) - (1 - \theta(X)) \frac{\partial W_a}{\partial H}(F, H) \] (10.56)

with \[ \theta(X) = 1 \text{ if } ||X|| \leq 1 \text{ and zero otherwise, for the deformation field } \chi(X) \text{ and the magnetic potential } \psi(X). \] In the above expressions, again, the effective free-energy function \( \overline{W} \) is given by
(10.44) for the case of magnetorheological elastomers wherein the fillers are spherical iron particles and by (10.49) for the case of magnetorheological elastomers wherein the fillers are spherical ferrofluid particles.

Figure 10.7: Contour plots of the component $F_{33}(\mathbf{X})$ of the deformation gradient over spherical specimens of magnetorheological elastomers containing $c = 0.20$ volume fraction of: (a)–(c) spherical iron filler particles and (d)–(f) spherical ferrofluid filler particles (with magnetization saturation $m_s = 0.3$ MA/m). The contours correspond to the remotely applied magnetic field $\mathbf{H}_\infty = H_\infty \mathbf{e}_3$ with $H_\infty = 0.11, 0.26, 1.00$ MA/m and, as implied by the argument $\mathbf{X}$ in $F_{33}(\mathbf{X})$, are shown over the undeformed configuration of the specimens. The color scale bars in each of the contour plots indicate the corresponding variation of $F_{33}(\mathbf{X})$ from its minimum to its maximum value.

For computational expediency, we seek to generate numerical solutions for the boundary-value problem (10.54) on a spatial domain of sufficiently large but finite extent, and not on $\mathbb{R}^3$ in its entirety. To this end, we find it convenient to consider a domain that consists of the specimen surrounded by an air-filled thick spherical shell that is subjected on its external surface to the affine deformation $\mathbf{x} = \mathbf{X}$ and the affine magnetic potential $\psi = -\mathbf{H}_\infty \cdot \mathbf{X}$; such a computational domain is schematically depicted in Fig. 10.6(a). A parametric study has revealed that a domain of this kind with initial outer radius twenty times larger than the initial radius of the specimen is large enough to accurately reproduce the solution of the boundary-value problem (10.54), at least for the choices of material and loading parameters of interest here. The study has also revealed that an initial shear modulus of the air that is three orders of magnitude smaller than that of the rubber
utilized for the specimen — namely, $G_a = 10^{-3} G$ — is small enough to be representative of $G_a = 0^+$. Now, given the axial symmetry of the problem around the direction of the applied magnetic field $H_\infty$, it proves useful to select the frame of reference so that $H_\infty = H_\infty e_3$. In this context, accurate numerical solutions can be efficiently generated by means of a conforming axisymmetric 7-node hybrid triangular finite element discretization on meshes comprising about 160,000 elements such as the one illustrated in Fig. 10.6(b). The representative solutions that we present next were generated based on such a discretization. All of these solutions correspond to specimens with the same rubber matrix, for which the function $\Psi$ is given by (10.5) with the parameters $G_1 = 0.1$ MPa, $G_2 = 0$ MPa, $\alpha_1 = \alpha_2 = 1$, and the same volume fraction of filler particles $c = 0.20$. For specimens wherein the fillers are spherical iron particles, the solutions correspond to the choice (10.8) for the function $S$ in (10.44) with the parameters $\mu_p = 100\mu_0$ and $m_s = 1.0$ MA/m. On the other hand, for specimens wherein the fillers are spherical ferrofluid particles, the solutions correspond to the choice (10.8) for the function $S$ in (10.49) with the parameters $\mu_p = 10\mu_0$ and $m_s = 0.1, 0.3, 0.5$ MA/m.

Figures 10.7(a)–(c) present contour plots in the $e_1$–$e_3$ plane of the component $F_{33}(X)$ of the local deformation gradient over the specimen of the magnetorheological elastomer with iron filler particles at the three values of the applied magnetic field $H_\infty = 0.11, 0.26$, and 1.00 MA/m. Figures 10.7(d)–(f) present the analogous contour plots for the specimen containing ferrofluid filler particles with magnetization saturation $m_s = 0.3$ MA/m. As already established by Brown (1966), an immediate
observation from these contour plots is that the deformation gradient in the specimens is *not* uniform, increasingly so for increasing values of the applied magnetic field. We notice in particular that the core of the magnetorheological elastomer with iron particles undergoes extension \((F_{33}(X) > 1)\), while its poles are under compression \((F_{33}(X) < 1)\). This extension/compression heterogeneity increases substantially and monotonically with the applied magnetic field. At the largest value \(H_\infty = 1.00\) MA/m of the applied magnetic field, when the specimen happens to have already reached a saturated state of magnetization, \(F_{33}(X) = 1.024\) at the center of the specimen while \(F_{33}(X) = 0.981\) at the poles. By contrast, the magnetorheological elastomer with ferrofluid particles, which exhibits a lesser degree of heterogeneity throughout the entire magnetic loading process, is under extension \((F_{33}(X) > 1)\) at every material point. The largest and smallest extensions occur at the center and at the poles of the specimen, respectively. For instance, at the largest value \(H_\infty = 1.00\) MA/m of the applied magnetic field, the specimen attains the value \(F_{33}(X) = 1.039\) at its center and \(F_{33}(X) = 1.028\) at its poles. By comparing these extremal values to those of the magnetorheological elastomer with iron particles, it is plain that the use of ferrofluid filler particles — in lieu of the more conventional iron filler particles — in magnetorheological elastomers can lead to significantly superior magnetostrictive properties.

To gain further quantitative insight into the enhancement of magnetostrictive properties afforded by ferrofluid particles, we plot in Fig. 10.8(a) the average deformation gradient \(\langle F_{33}(X) \rangle = (\int_{R^3} \theta(X)dX)^{-1} \int_{R^3} \theta(X)F_{33}(X)dX\), or average magnetostriction, as a function of the applied magnetic field \(H_\infty\) for the two specimens shown in Fig. 10.7. As expected from the preceding observations about the local deformation, the plot shows that the specimen with ferrofluid filler particles exhibits an average magnetostriction of extension \((\langle F_{33}(X) \rangle > 1)\) that is significantly larger than that of the specimen with iron filler particles; interestingly, the average magnetostriction of the latter is also of extension. Furthermore, we remark that the average magnetostriction saturates in both specimens as the magnetic field increases, but that in the specimen with ferrofluid filler particles the saturation is reached at a significantly smaller value of the applied magnetic field. A parametric study varying the magnetization saturation parameter \(m_s\) of the ferrofluid has indicated that the saturated value of the average magnetostriction, \(\langle F_{33}(X) \rangle_{\text{sat}}\), can be increased significantly by increasing the value of \(m_s\). This point is illustrated by Fig. 10.8(b), where analogous results to that presented in Fig. 10.8(a) are plotted for specimens containing ferrofluid filler particles with \(m_s = 0.1, 0.3,\) and 0.5.
MA/m. These encouraging sample results are supportive of further investigations of magnetorheological elastomers containing ferrofluid filler particles.

We close this section, for completeness, by presenting in Fig. 10.9 the contour plots of the component $H_3(X)$ of the Lagrangian magnetic field over the same two spherical specimens discussed in Fig. 10.7. Similar to the deformation fields, the magnetic fields are heterogeneous, more so for the specimen with iron filler particles.

![Figure 10.9: Contour plots of the component $H_3(X)$ of the Lagrangian magnetic field from the same simulations as in Fig. 10.7 over spherical specimens of magnetorheological elastomers containing $c = 0.20$ volume fraction of: (a)–(c) spherical iron filler particles and (d)–(f) spherical ferrofluid filler particles (with magnetization saturation $m_s = 0.3$ MA/m). The contours correspond to the remotely applied magnetic field $H_\infty = H_\infty e_3$ with $H_\infty = 0.11, 0.26, 1.00$ MA/m and, as implied by the argument $X$ in $H_3(X)$, are shown over the undeformed configuration of the specimens. The color scale bars in each of the contour plots indicate the corresponding variation of $H_3(X)$ from its minimum to its maximum value.](image)

**10.6 Comparison with experiments and final comments**

Over the last decade, numerous experimental investigations have been devoted to probe the magnetoeleastic behavior of magnetorheological elastomers at finite deformations and finite magnetic fields. Most of them have focused on probing the effects of the content and the distribution of the underlying iron filler particles (see, e.g., Varga et al., 2005; Guan et al., 2008; Diguet, 2010; Danas et
al., 2012). There are also experiments which have investigated other effects, such as the presence of pores (Bednarek, 2006; Ju et al., 2012), the interfacial bonding between the rubber matrix and the iron filler particles (Wang et al., 2006), and the type of rubber matrix (Ge et al., 2013). A common challenge in all of these experimental works is that the deformation and magnetic fields experienced by the specimens are, in general, not homogeneous. This makes it difficult to quantify the actual constitutive magnetoelastic behavior of the magnetorheological elastomer being tested, as the response measured experimentally contains a structural contribution dependent on the geometry of the specimen. Equipped with the general result (10.16) for the effective free energy of isotropic magnetorheological elastomers and the finite-element framework outlined in Section 10.5, we are now in a position to examine experimental results by carrying out simulations of the experiments accounting directly for the geometry of the specimen at hand, as well as for the constitutive behavior of the rubber matrix and the constitutive behavior, volume fraction, and (isotropic) distribution of the iron filler particles. In this section, by way of an example, we examine experimental results due to Diguet (2010); see also Diguet et al. (2009) and Diguet et al. (2010). This author fabricated cm-scale specimens of spherical and cylindrical shape comprised of a soft silicone rubber isotropically filled with spherical iron particles of about 5 \( \mu \)m diameter at several volume fractions \( c \in [0.05, 0.35] \).

Among the various experiments that he conducted, we consider here those aimed at investigating magnetostriction. These consisted in subjecting the specimens to a roughly uniaxial magnetic field of increasing magnitude and monitoring their deformed geometry as a function of the applied field.

In order to reproduce theoretically the magnetostriction experiments by Diguet (2010), we find it useful to describe the initial geometry of the specimens with the family of domains

\[
\Omega = \left\{ \mathbf{X} \in \mathbb{R}^3 : \left( \frac{X_1^2}{L_1^2} + \frac{X_2^2}{L_2^2} \right)^{\frac{1}{1-k}} + \left( \frac{X_3^2}{L_3^2} \right)^{\frac{1}{1-k}} \leq 1 \right\},
\]

(10.57)

where \( 0 \leq k < 1, L_1, L_2, L_3 > 0 \) are real-valued parameters. Indeed, a spherical specimen of radius \( A \) corresponds to setting \( k = 0 \) and \( L_1 = L_2 = L_3 = A \), while a cylindrical specimen of radius \( A \) and height \( B \) corresponds to setting \( k = 1^{-} \), \( L_1 = L_2 = A, L_3 = B/2 \). Cylindrical specimens with round corners, or fillets, can also be described with the parametrization (10.57) by choosing a value of \( k \) sufficiently smaller than 1. Note that the center of the specimens, as described by (10.57), has been tacitly chosen as the origin of the frame of reference.

The nonlinear elastic behavior of the soft silicone rubber utilized to make the specimens was not reported. Diguet (2010) did report, however, that it is a bi-compound silicone from Dalbe featuring a Young’s modulus and Poisson’s ratio of about 100 kPa and 0.5. In the simulations that follow, given
this limited information, we assume that the nonlinear elastic behavior of the silicone rubber matrix is approximately characterized by the free energy (10.1) with the function $\Psi$ given by (10.5) and one of the two sets of material parameters listed in Table 10.1. These parameters correspond to the presumably similar bi-compound sylgard 184 silicone rubber from Dow Corning with ratios 30:1 and 45:1 of PDMS base to curing agent; see Section 3 in Poulain et al. (2017). The Young’s modulus of the 30:1 silicone is $3(G_1 + G_2) = 151.47$ kPa and that of the 45:1 silicone $3(G_1 + G_2) = 44.31$ kPa. These values are indeed comparable to the one indicated by Diguet (2010).

<table>
<thead>
<tr>
<th>composition</th>
<th>$\alpha_1$</th>
<th>$\alpha_2$</th>
<th>$G_1$ (kPa)</th>
<th>$G_2$ (kPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30:1</td>
<td>-1.02103</td>
<td>1.39107</td>
<td>18.57</td>
<td>31.92</td>
</tr>
<tr>
<td>45:1</td>
<td>-1.10010</td>
<td>1.45673</td>
<td>5.22</td>
<td>9.55</td>
</tr>
</tbody>
</table>

Table 10.1: Material parameters in the function (10.5) for two compositions — 30:1 and 45:1 ratios of PDMS base to curing agent — of the silicone rubber matrix.

Based on X-ray analyses, the filler particles utilized to make the specimens were reported to be 98.7% iron. For definiteness, we choose (10.8) with permeability $\mu_p = 100\mu_0$ and magnetization saturation $m_s = 1.66$ MA/m for the function $S$ in (10.16) to describe their constitutive behavior. The fabrication process led to the clustering of filler particles but no information was provided about the sizes and shapes of these. In the simulations, as a first reasonable approximation, we assume that they are roughly spherical, firmly bonded to the silicone rubber matrix, and thus make use of the specialized result (10.44) for the effective free-energy function $\overline{W}$.

The experiments were conducted in between two electromagnets separated by a 4-cm gap. In the simulations, in order to avoid having to explicitly model the electromagnets (which is admittedly a non-trivial task left for a future work), we assume that these are infinitely far apart and that the applied magnetic field at infinity is uniform and uniaxial. More precisely, much like in the preceding section, we consider applied uniform magnetic fields of the form $H_\infty = H_\infty e_3$ and follow the same computational strategy to numerically solve the resulting boundary-value problem.

Figures 10.10 and 10.11 show results from the simulations of two experiments on: (a)–(c) a spherical specimen of initial radius $A = 0.98$ cm and volume fraction of iron filler particles $c = 0.28$ and (d)–(f) a cylindrical specimen of initial radius $A = 0.59$ cm, initial height $B = 0.73$ cm, and particle volume fraction $c = 0.20$. For definiteness, the corners in the cylindrical specimen are taken to be described by the value of $k = 0.95$ in the parametrization (10.57); larger values of $k$ were checked to render essentially the same results. Both sets of plots pertain to simulations with
Figure 10.10: Contour plots of the component \( F_{33}(X) \) of the deformation gradient from the simulations of two experiments on: (a)–(c) a spherical specimen of initial radius \( A = 0.98 \) cm and volume fraction of iron filler particles \( \phi = 0.28 \) and (d)–(f) a cylindrical specimen of initial radius \( A = 0.59 \) cm, initial height \( B = 0.73 \) cm, and volume fraction \( \phi = 0.20 \) of iron particles. The results correspond to the 30:1 silicone rubber matrix and the remotely applied magnetic field \( H_{\infty} = H_{\infty} e_3 \) for the three values \( H_{\infty} = 0.16, 0.41, \) and 1.00 MA/m. The color scale bars in each of the contour plots indicate the corresponding variation of \( F_{33}(X) \) from its minimum to its maximum value.

the 30:1 silicone rubber matrix. The results display the contour plots in the \( e_1-e_3 \) plane of the components \( F_{33}(X) \) and \( H_3(X) \) of the local deformation gradient and Lagrangian magnetic field over the specimens at the applied magnetic fields \( H_{\infty} = 0.16, 0.41, \) and 1.00 MA/m. The last of these three values, \( H_{\infty} = 1.00 \) MA/m, was selected so that it is large enough to correspond to saturated states of magnetization in both specimens.

A quick glance at the contours displayed in Fig. 10.10 suffices to recognize that the deformation gradient is highly heterogeneous in both specimens. Consistent with the results of Section 10.5, the spherical specimen undergoes extension \( (F_{33}(X) > 1) \) in its core and compression \( (F_{33}(X) < 1) \) around its poles, featuring, for instance, maximum and minimum values of \( F_{33}(X) = 1.124 \) and \( F_{33}(X) = 0.908 \) at the applied magnetic field \( H_{\infty} = 1.00 \) MA/m. On the other hand, the cylindrical specimen undergoes compression \( (F_{33}(X) < 1) \) throughout its core and at its corners, while, rather interestingly, the regions neighboring the compressed corners are under extension \( (F_{33}(X) > 1) \). The maximum and minimum values attained in this case at \( H_{\infty} = 1.00 \) MA/m are \( F_{33}(X) = 1.038 \)
10. Magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber

Figure 10.11: Contour plots of the component $H_3(X)$ of the Lagrangian magnetic field from the same simulations as in Fig. 10.11 of two experiments on: (a)–(c) a spherical specimen of initial radius $A = 0.98$ cm and volume fraction of iron filler particles $c = 0.28$ and (d)–(f) a cylindrical specimen of initial radius $A = 0.59$ cm, initial height $B = 0.73$ cm, and volume fraction $c = 0.20$ of iron particles. The results correspond to the 30:1 silicone rubber matrix and the remotely applied magnetic field $H_\infty = H_\infty e_3$ for the three values $H_\infty = 0.16$, 0.41, and 1.00 MA/m.

and $F_{33}(X) = 0.953$. A large number of simulations, not reported here, have shown that the above-outlined qualitative features of the local deformation are largely insensitive to the size of the specimens (that is, the radius $A$ for spherical specimens and the radius $A$ and height $B$ for cylindrical ones) and to the volume fraction $c$ of iron filler particles that they contain.

The contours displayed in Fig. 10.11 show that the Lagrangian magnetic field is also highly heterogeneous in both specimens. For the spherical specimen, we notice in particular that the local magnetic field component $H_3(X)$ is largest around the core. For the cylindrical specimen, on the other hand, $H_3(X)$ is largest along the circumference. For the spherical specimen, we also remark that the corresponding Eulerian magnetic field $h_3(x)$ is drastically more homogeneous over the specimen, presumably because its deformed shape is not far from ellipsoidal. This point — which has practical implications, for instance, in allowing for direct experimental measurements of magnetization at finite deformations — is illustrated by the contour plots of $h_3(x)$ displayed in Fig. 10.12(a). By contrast, as illustrated in Fig. 10.12(b), the cylindrical specimen exhibits an Eulerian magnetic field that is as highly heterogeneous as its Lagrangian counterpart. In connection with this
interesting feature, we further remark that both the first Piola-Kirchhoff stress component $S_{33}(X)$ and the Cauchy stress component $T_{33}(x)$ are markedly heterogeneous in the spherical as well as in the cylindrical specimens. This point is illustrated by the contour plots displayed in Fig. 10.13.

Having gained insight into the local deformation and magnetic fields within the specimens, we now turn to examining a macroscopic measure of their magnetostriction at saturated states of magnetization for direct comparison with the experimental measurements of Diguet (2010). In particular, this author reported the “overall magnetostriction stretch” $\lambda$ between the poles of the spherical specimens and between the top and bottom circumferences of the cylindrical specimens, as defined in the schematics of Fig. 10.14. We emphasize that such a macroscopic measure of magnetostriction does not correspond to the average magnetostriction $\langle F_{33}(X) \rangle$ over the specimens, but to an approximation of it. The table in part (c) of Fig. 10.14 reports the saturated overall magnetostriction stretch $\lambda$ for a spherical specimen of initial radius $A = 0.98$ cm containing $c = 0.28$ volume fraction of iron filler particles. Figure 10.14(f) displays the values of $\lambda$ for cylindrical specimens of initial radius $A = 0.59$ cm and initial height $B = 0.73$ cm, as a function of the volume fraction of iron filler particles $c$. All the simulation results in both parts (c) and (f) are shown for the two compositions 30:1 and 45:1 of the silicone rubber matrix. In the latter part, the solid lines denote results from the simulations, while the solid triangles stand for the experimental data.

In qualitative agreement with the experimental results, the simulations indicate that the overall magnetostriction of all the specimens, as characterized by the stretch $\lambda$, is of extension ($\lambda > 1$).
We emphasize that this behavior is non-trivial as large portions of the specimens are locally under compression. Quantitatively, the agreement between the simulations and the experimental result for the spherical specimen is also admittedly good, especially for the stiffer composition 30:1 of the silicone rubber matrix. The same is not true for the cylindrical specimens, for which there are significant quantitative differences between the simulations and the experimental data.

In an attempt to pinpoint the source of the difference between the theoretical and the experimental results for the cylindrical specimens, we carried out a number of simulations where we varied the stiffness of the silicone rubber matrix, accounted for the presence of pores possibly due to an incomplete degassing in the fabrication process of the specimens, and varied the shape of the underlying filler particles to non-spherical. None of these changes led to satisfactory quantitative agreement with the experiments. Given this evidence, we conjecture that the magnetic fields applied in the experiments, as generated within a 4-cm gap between two electromagnets, cannot accurately be approximated as remotely uniform and uniaxial. Instead, one would have to account for the explicit presence of the electromagnets in the simulations in order to theoretically reproduce the actual magnetic fields that they generate in the presence of the specimens. Some results in this direction can be found, for instance, in the works of Salas and Bustamante (2015) and Pössinger (2015).

A few words from an applications perspective are in order to conclude this chapter. The above sample results have made it plain that the overall magnetostriction of specimens made up of isotropic magnetorheological elastomers with iron filler particles is strongly dependent on the elasticity of the underlying rubber matrix, the volume fraction of the particles, as well as on the geometry of the specimens. In particular, sizable overall magnetostrictions (in the order of 10% uniaxial strains) can be achieved using a sufficiently soft rubber matrix (featuring initial shear moduli in the tens of kPAs) filled with a sufficiently large volume fraction of iron particles (in the range of $c = 0.20$ to $c = 0.35$). The strong dependence of the overall magnetostriction on the specimen geometry calls for attempts at designing magnetostrictive devices based on magnetorheological elastomers to be approached as structural problems, and not simply as a materials design problems.
10. Magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber

Figure 10.13: Contour plots of the components $S_{33}(X)$ and $T_{33}(x)$ of the first Piola-Kirchhoff and Cauchy stress tensors from the same simulations as in Fig. 10.10 of two experiments on: (a)–(b) a spherical specimen of initial radius $A = 0.98$ cm and volume fraction of iron filler particles $c = 0.28$ and (c)–(d) a cylindrical specimen of initial radius $A = 0.59$ cm, initial height $B = 0.73$ cm, and volume fraction $c = 0.20$ of iron particles. The contours correspond to the remotely applied magnetic field $H_\infty = 1.00$ MA/m and, as implied by the arguments $X$ and $x$ in $S_{33}(X)$ and $T_{33}(x)$, are shown over the undeformed configuration (parts (a) and (c)) and over the deformed configuration (parts (b) and (d)) of the specimens.
Figure 10.14: Schematics of a spherical specimen of initial radius $A$ in (a) its initial configuration and (b) its deformed configuration at a saturated state of magnetization indicating the two material points at the poles utilized to compute the overall magnetostriction stretch $\lambda = a/A$. Schematics of a cylindrical specimen of initial radius $A$ and initial height $B$ in (d) its initial configuration and (e) its deformed configuration at a saturated state of magnetization indicating the two material points at the circumferences utilized to compute the overall magnetostriction stretch $\lambda = b/B$. The table in part (c) reports the overall magnetostriction stretch $\lambda$ for a spherical specimen of initial radius $A = 0.98$ cm with particle volume fraction $c = 0.28$. The plots in part (f) show $\lambda$ as a function of the volume fraction of iron filler particles $c$ for cylindrical specimens of initial radius $A = 0.59$ cm and initial height $B = 0.73$ cm. All results from the simulations are shown for the two compositions 30:1 and 45:1 of the silicone rubber matrix.
Closure

Il faut que les jeunes gens s’habituent à voir par eux-mêmes et sans demander avis, la différence du beau et du laid.

– Auguste Renoir, Grammaire, 1883–1884

An expert is a person who has found out by his own painful experience all the mistakes that one can make in a very narrow field.

– Niels Bohr, quoted by Edward Teller in LIFE magazine, 1954

This dissertation has put forth analytical and numerical homogenization methods to determine the macroscopic elastic dielectric response of dielectric elastomer composite materials. Specifically, rigorous homogenization solutions in two and three space dimensions for dielectric elastomer composites with general (possibly anisotropic) classes of two-phase particulate microstructures were constructed in the asymptotic context of small deformations and moderate electric fields. These asymptotic solutions proved later to be essential building blocks for the development of corresponding homogenization solutions for finite deformations and finite electric fields. The resulting general variational homogenization solution applies to isotropic non-Gaussian dielectric elastomers filled with isotropic suspensions of nonlinear elastic dielectric particles that may exhibit polarization saturation. This approximate solution is exact by construction in the limit of small deformations and moderate electric fields. For finite deformations and finite electric fields, its accuracy was assessed by direct comparisons with full-field hybrid finite-element simulations, as well as with numerical solutions generated via a new WENO finite-difference scheme developed specifically for this class of problems.
Glaring disagreements between the theoretical response predicted from these methods for two-phase particulate composites free from any space charges and a number of experimental results have made it plain, however, that this initial microscopic description of dielectric elastomer composites is fundamentally incomplete, especially for cases involving stiff filler particles. Consequently, the presence of space charges that oscillate rapidly at the length scale of the microstructure was incorporated into the microscopic description of these dielectric elastomer composites. The resulting homogenized equations revealed that the presence of (passive or active) space charges within elastic dielectric composites can have a significant and even dominant effect on their macroscopic response, possibly leading to extreme behaviors ranging from unusually large permittivities and electrostriction coefficients to metamaterial-type properties featuring negative permittivities. These results suggest a promising strategy to design deformable dielectric composites — such as electrets and dielectric elastomer composites — with exceptional electromechanical properties.

The analytical and numerical homogenization methods put forth for the macroscopic elastic dielectric response of dielectric elastomer composites were also utilized to put forth, within the mathematically analogous setting of magnetoelastostatics, an approximate analytical solution for the effective free-energy function describing the homogenized (or macroscopic) magnetoelastic response of magnetorheological elastomers comprised of non-Gaussian rubbers filled with isotropic suspensions of either iron or ferrofluid particles. It was found that magnetorheological elastomers filled with ferrofluid particles can exhibit magnetostrictive capabilities far superior to those of magnetorheological elastomers filled with iron particles. The results also revealed that the deformation and magnetic fields are highly heterogeneous within the specimens and strongly dependent on the shape of these, specially for magnetorheological elastomers filled with iron particles. From an applications perspective, this evidence indicates that attempts at designing magnetostrictive devices based on magnetorheological elastomers need to be approached, in general, as structural problems, and not simply as materials design problems.

To close, we outline the challenges — in particular at finite deformations and finite electric fields — of the microscopic description that views dielectric elastomer composites as N-phase particulate composites containing source terms (i.e., space charges) that spatially vary at the length scale of microstructure and how they could be addressed. We also describe a few directions in which the results presented in this work could be extended.

In the absence of space charges, we have already demonstrated (see the closing paragraphs of
Chapter 7) that the proposed closed-form approximate solution (7.26) provides an accurate solution for a three-phase particulate ideal elastic dielectric composite wherein the filler particles are bonded through finite-size interphases to the dielectric elastomer matrix. Starting with (7.26) and in the same spirit of Chapter 8, it would not be difficult to extend the solution (8.47) to this class of dielectric elastomer composites in order to account for the non-Gaussian behavior of the matrix, the possible polarization-saturation of the particles, and the presence of interphases. Based on the results of Goudarzi et al. (2015) for the pure mechanical case ($\mathbf{E} = \mathbf{0}$), it is expected that such an extension could provide a fairly accurate description of dielectric elastomer composites with interphases.

The incorporation of interphasial space charges in the context of finite deformations and finite electric fields appears to be a much more delicate endeavor. Through minor modifications to the framework presented in Section 8.3, FE solutions can be readily obtained for the relevant governing equations; see Figs. 8.9(a) and 9.6(b). In this regard, the FE method shall prove to be a formidable tool to provide insight into the macroscopic elastic dielectric behavior of this class of dielectric elastomer composites comprising interphasial charges. From the analytical standpoint, however, the presence of source terms that vary at the microscopic length scale calls for the revision of the definition (2.14) of the macrovariables. The results in Sections 9.2 and 9.3 of Chapter 9 (see also the work of Lopez-Pamies et al. (2014) in the context of linear dielectrics) combined with the above-discussed FE solutions could prove useful to derive (at least formally) expressions for the macrovariables in finite deformations and finite electric fields. Then, it might not be difficult to extend the results (4.23) and (4.48) for the macroscopic elastic dielectric response — in the limit of small deformations and moderate electric fields — of dielectric elastomer composites comprising the differential microstructures discussed in Chapter 4, now comprising interphasial regions with different mechanical and/or physical properties than the matrix phase, and space charges. Following the strategies employed in Chapters 7 and 8, equipped with the effective material parameters that characterize their macroscopic response in the small-deformation limit, it would then be interesting to assess the accuracy of the solutions (7.26) and the above-described extension of (8.47) for these classes of dielectric elastomer composites comprising interphasial charges and, finally confront them with the experimental results.

Finally, we recall here that irreversible (or dissipative) mechanical and/or dielectric phenomena such as viscoelasticity, fracture, dielectric loss, electric breakdown were not accounted for in the work presented in this document. Accounting for these phenomena at finite deformations and finite
electric fields, a challenging task already for homogeneous materials, is likely to require ultimately the use and/or development of further analytical and numerical frameworks.
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Bibliography


A

The F and D formulation

The microscopic description. At several passages in this document, it proves useful to treat the Lagrangian electric displacement $\mathbf{D}$ as the independent local electric variable instead of the Lagrangian electric field $\mathbf{E}$. Among several possibilities, this can be readily accomplished with help of suitable partial Legendre transforms. Indeed, for the case of interest in this work when $W(\mathbf{X}, \mathbf{F}, \mathbf{E})$ is concave in $\mathbf{E}$, the partial Legendre transform

$$W^*(\mathbf{X}, \mathbf{F}, \mathbf{D}) = \sup_{\mathbf{E}} \{ \mathbf{D} \cdot \mathbf{E} + W(\mathbf{X}, \mathbf{F}, \mathbf{E}) \},$$

which physically corresponds to the local Helmholtz free energy, allows us to write formally the local constitutive relation for the dielectric elastomer composite in the form

$$\mathbf{S} = \frac{\partial W^*}{\partial \mathbf{F}}(\mathbf{X}, \mathbf{F}, \mathbf{D}) \quad \text{and} \quad \mathbf{E} = \frac{\partial W^*}{\partial \mathbf{D}}(\mathbf{X}, \mathbf{F}, \mathbf{D}),$$

(A.2)

where now $\mathbf{D}$ plays the role of independent electric variable. Given the above-described functional properties of the local free energy $W(\mathbf{X}, \mathbf{F}, \mathbf{E})$, it is not difficult to show that the local Helmholtz free energy $W^*(\mathbf{X}, \mathbf{F}, \mathbf{D})$ is an objective function of $\mathbf{F}$ and an even and objective function of $\mathbf{D}$, namely, $W^*(\mathbf{X}, \mathbf{F}, \mathbf{D}) = W^*(\mathbf{X}, \mathbf{QF}, \mathbf{D}) = W^*(\mathbf{X}, \mathbf{F}, -\mathbf{D})$.

The macroscopic response. Similarly, at several passages in this document, it will prove useful to treat the macroscopic Lagrangian electric displacement $\bar{\mathbf{D}}$ as the independent macroscopic electric variable instead of the macroscopic Lagrangian electric field $\bar{\mathbf{E}}$. For the case when $\bar{W}(\mathbf{F}, \bar{\mathbf{E}}, c)$ is concave in $\bar{\mathbf{E}}$, the partial Legendre transform

$$\bar{W}^*(\mathbf{F}, \bar{\mathbf{D}}, c) = \sup_{\bar{\mathbf{E}}} \{ \bar{\mathbf{D}} \cdot \bar{\mathbf{E}} + \bar{W}(\mathbf{F}, \bar{\mathbf{E}}, c) \}$$

(A.3)
allows us to write formally the macroscopic constitutive relation for the dielectric elastomer composite in the form
\[
S = \frac{\partial W^*}{\partial F} (F, D, c) \quad \text{and} \quad E = \frac{\partial W^*}{\partial D} (F, D, c)
\]  
(A.4)
Differential coated microstructures in the small-deformation limit: additional constants

We report in this appendix the various additional constants entering in the derivation of the macroscopic elastic dielectric behavior of differential coated microstructures in the small-deformation limit presented in Chapter 4.

B.1 Differential coated sphere assemblage

B.1.1 The constants $A_1, A_3, B_1, B_2, B_3, B_4$

The six constants $A_1, A_3, B_1, B_2, B_3, B_4$ in the functions (4.15) and (4.16) are defined by the continuity of the fields $\Gamma_{rkl}$ and $L_{ijrs} \Gamma_{rkl,s} X_j$ across the two material interfaces at $R = R_p, R_m$, and by the boundary condition at infinity where $\Gamma_{ikl} = \delta_{ik} \delta_{jl} X_j$. These conditions lead to six linear
equations for the six constants, namely,

\[
E_1 = -A_1 - c^{2/3} A_3 + B_1 + \frac{B_2}{c^{5/3}} + c^{2/3} B_3 + \frac{B_4}{c} = 0,
\]

\[
E_2 = \frac{2(6\kappa_\mu + 17\mu_p)A_3}{15\kappa_\mu + 11\mu_p} - \frac{5B_2}{c^{7/3}} - \frac{2(6\kappa_\mu + 17\mu)B_3}{15\kappa_\mu + 11\mu} + \frac{(3\kappa + \mu)B_4}{c^{5/3}} = 0,
\]

\[
E_3 = -\mu_p A_1 - \frac{63c^{2/3}\mu_p^2 A_3}{19(15\kappa_\mu + 11\mu_p)} + \mu B_1 + \frac{84\mu B_2}{19c^{5/3}} + \frac{63c^{2/3}\mu^2 B_3}{19(15\kappa_\mu + 11\mu)} - \frac{135\kappa + 64\mu)B_4}{38c} = 0,
\]

\[
E_4 = \frac{\mu_p(57c_\mu + 4\mu_p)A_3}{15\kappa_\mu + 11\mu_p} + \frac{20\mu B_2}{c^{7/3}} - \frac{\mu(57\kappa + 4\mu)B_3}{15\kappa_\mu + 11\mu} - \frac{4(3\kappa + \mu)B_4}{c^{5/3}} = 0,
\]

\[
E_5 = \frac{6\mu(6\kappa + 2\mu)}{[\mu(9\kappa_\mu + 8\mu)] + 1} B_1 + \left(1 - \frac{\mu}{\mu}\right) B_2 + \left[\frac{28\mu(3\kappa + \mu)}{(15\kappa_\mu + 11\mu)(9\kappa_\mu + 8\mu)} + \frac{3\mu(15\kappa + 4\mu)}{60\kappa_\mu + 44\mu} + 1\right] B_3 + \frac{5(3\kappa + 4\mu)}{2(9\kappa_\mu + 8\mu)} = 0,
\]

\[
E_6 = 5\left(\frac{\mu}{\mu} - 1\right) B_2 - \frac{[\kappa(5\kappa + 6\mu)]}{60\kappa_\mu + 44\mu} - \frac{15\kappa + 64\mu)B_4}{\mu_\mu} = 0.
\]

While it is simple matter to compute the unique solution to this system of linear equations for \( A_1, A_3, B_1, B_2, B_3, B_4 \), we do not report it here because of the bulkiness of the final expressions.

**B.1.2 Expressions for \( q_1, q_2, q_3 \)**

The expressions for \( q_1, q_2, q_3 \) in the result (4.23) for the effective shear modulus \( \tilde{\mu} \) are given by

\[
q_1 = 6c^{10/3}\eta_1 \left[ 2 + \frac{\kappa}{\mu} \right] - 126c^{5/3}\eta_3 \left( 1 + \frac{3\kappa}{\mu} \right)^2 + c^{7/3}\eta_2 + 225c\eta_3 \left( \frac{3\kappa^2}{\mu^2} + \frac{4\kappa}{\mu} + 3 \right) - \frac{3\kappa_3}{\mu_p - \mu} \left[ 6\mu_2 \left( \frac{\kappa}{\mu} + 2 \right) + \frac{9\kappa}{\mu} + 8 \right],
\]

\[
q_2 = c^{10/3}\eta_1 \left[ 4 - \frac{3\kappa}{\mu} \right] - 252c^{5/3}\eta_3 \left( 3\frac{\kappa}{\mu} + 1 \right)^2 + 2c^{7/3}\eta_2 + \frac{75}{4}c\eta_3 \left( \frac{3\kappa}{\mu} - 2 \right) \left( \frac{15\kappa}{\mu} + 8 \right) + \frac{9\kappa_3}{8(\mu_p - \mu)} \left[ 6\mu_2 \left( \frac{\kappa}{\mu} + 2 \right) + \frac{9\kappa}{\mu} + 8 \right],
\]

\[
q_3 = c^{10/3}\eta_1 \left[ 8 + \frac{9\kappa}{\mu} \right] + 126c^{5/3}\eta_3 \left( 3\frac{\kappa}{\mu} + 1 \right)^2 - c^{7/3}\eta_2 - \frac{75}{8}c\eta_3 \left( \frac{81\kappa^2}{\mu^2} + \frac{60\kappa}{\mu} + 8 \right) - \frac{3\kappa_3}{8(\mu_p - \mu)} \left[ 6\mu_2 \left( \frac{\kappa}{\mu} + 2 \right) + \frac{9\kappa}{\mu} + 8 \right].
\]
where

\begin{align*}
\eta_1 &= 3 \left(1 - \frac{\mu_p}{\mu}\right) \left\{ \frac{3\kappa}{\mu} \left[ \frac{114\kappa_p}{\mu} \left(1 - \frac{\mu_p}{\mu}\right) + \frac{\mu_p}{\mu} \left(323 - \frac{8\mu_p}{\mu}\right) \right] + \frac{3\kappa_p}{\mu} \left(8 - \frac{323\mu_p}{\mu}\right) + \frac{68\mu_p}{\mu} \left(1 - \frac{\mu_p}{\mu}\right) \right\}, \\
\eta_2 &= 225 \left\{ \frac{3\kappa^2}{\mu^2} \left[ \frac{54\kappa_p}{\mu} - \frac{\mu_p^2}{\mu^2} \left(57\kappa_p \mu + 86\right) + \frac{3\mu_p}{\mu} \left(\kappa_p \mu + 51\right) - 4\mu_p^3 \right] + \frac{\kappa}{\mu} \left[ \frac{120\kappa_p}{\mu} - \frac{12\mu_p^2}{\mu^2} \left(19\kappa_p \mu + 6\right) + \frac{3\mu_p}{\mu} \left(340 - \frac{81\kappa_p}{\mu}\right) - \frac{16\mu_p^3}{\mu^3} \right] + \frac{\kappa_p}{\mu} \left[ 16 - \frac{171\mu_p^2}{\mu^2} - \frac{9\kappa_p}{\mu} \right] + \frac{4\mu_p}{\mu} \left(1 - \frac{\mu_p}{\mu}\right) \left(\frac{9\kappa_p}{\mu} + 34\right) \right\}, \\
\eta_3 &= \left(1 - \frac{\mu_p}{\mu}\right) \left[ \frac{\kappa_p}{\mu} \left(\frac{57\mu_p}{\mu} + 48\right) + \frac{4\mu_p}{\mu} \left(\frac{\mu_p}{\mu} + 34\right) \right].
\end{align*}

(B.3)

B.2 Differential coated cylinder assemblage

B.2.1 The constants $A_1, A_3, B_1, B_2, B_3, B_4$

The six constants $A_1, A_3, B_1, B_2, B_3, B_4$ in the functions (4.34) and (4.35) are defined by the solution of the following system of six linear algebraic equations:

\begin{align*}
E_1 &= A_1 + A_3 c - B_1 - \frac{B_2}{c^2} - B_3 c - \frac{B_4}{c} = 0, \\
E_2 &= A_3 \frac{\lambda_p + 3\mu_p}{2\lambda_p + 3\mu_p} - 2B_2 - B_3 \frac{\lambda + 3\mu}{2\lambda + 3\mu} + B_4 \frac{\lambda + \mu}{c^2} = 0, \\
E_3 &= A_1 c^2 \mu_p - B_1 c^2 \mu - 3B_2 \mu + 2B_4 c(\lambda + \mu) = 0, \\
E_4 &= A_3 \frac{c^3 \mu_p (\lambda + \mu_p)}{2\lambda_p + 3\mu_p} + 2B_2 \mu - B_3 \frac{c^3 \mu (\lambda + \mu)}{2\lambda + 3\mu} - B_4 c(\lambda + \mu) = 0, \\
E_5 &= B_1 \left(1 + \frac{4\mu}{c_L} + \frac{2\mu}{e_L}\right) + B_2 \left(1 - \frac{2\mu}{e_L}\right) + B_3 \left(1 + \frac{2\mu (2\tilde{c}_L + 3\bar{e}_L)(\lambda + \mu)}{c_L e_L (2\lambda + 3\mu)}\right) + B_4 \left(1 - \frac{2(\lambda + \mu)}{c_L}ight) - \left(1 + \frac{\tilde{e}_L}{c_L}\right) = 0, \\
E_6 &= 2B_2 \left(1 - \frac{2\mu}{e_L}\right) + \frac{B_3}{2\lambda + 3\mu} \left(\lambda + 3\mu + \frac{2\mu (\lambda + \mu)}{e_L}\right) - B_4 \left(1 - \frac{2\mu}{e_L}\right) \left(1 + \frac{\lambda}{\mu}\right) = 0. \quad (B.4)
\end{align*}

These equations stem from the continuity of the fields $\Gamma_{rkl}$ and $L_{ijrs} \Gamma_{rkl,s} X_j$ across the two material interfaces at $R = R_p, 1$, and from the boundary condition at infinity where $\Gamma_{ikl} = \delta_{ik} \delta_{jl} X_j$. While it is a simple matter to compute the unique solution to this system of equations, we do not report it here because of the bulkiness of the final expressions.
B.2.2 Expressions for $q_1$, $q_2$, $q_3$

The expressions for $q_1$, $q_2$, $q_3$ in the result (4.48) for the effective elastic constant $\tilde{e}_{L}^{DCC}$ are given by

\begin{align*}
q_1 &= (\lambda + 3\mu)\tau_1 c^4 - 4\tau_2 c^3 + (\lambda + 3\mu)\tau_3\tau_4 + 2c\tau_3(\mu_p - \mu)[3c(\lambda + \mu)^2 - 2(\lambda^2 + 3\lambda\mu + 3\mu^2)]], \\
q_2 &= \mu\tau_1 c^4 - 4\tau_2 c^3 + \mu\tau_3\tau_4 + 2c\tau_3(\mu_p - \mu)(\lambda + \mu)[3c(\lambda + \mu) - \lambda], \\
q_3 &= (\lambda + \mu)\tau_1 c^4 + 4\tau_2 c^3 + (\lambda + \mu)\tau_3\tau_4 + 2c(2 - 3c)(\mu_p - \mu)(\lambda + \mu)^2\tau_3, \quad (B.5)
\end{align*}

where

\begin{align*}
\tau_1 &= (\mu_p - \mu)\left\{\mu_p[\lambda(\lambda_p - 3\mu) + 3\mu(\lambda_p - \mu)] - \lambda_p\mu(\lambda + \mu) + \mu_p^2(\lambda + 3\mu)\right\}, \\
\tau_2 &= \mu_p^2\left[\lambda^2(\lambda_p + \mu) + \lambda\mu(3\lambda_p + \mu) + 3\lambda_p\mu^2\right] + \mu_p^3\left(\lambda^2 + 3\lambda\mu + 3\mu^2\right) - (\lambda_p + 3\mu_p)(\lambda + \mu)^2\mu^2, \\
\tau_3 &= \lambda_p(\mu + \mu_p) + \mu_p(3\mu + \mu_p), \\
\tau_4 &= \lambda(\mu + \mu_p) + \mu(\mu + 3\mu_p). \quad (B.6)
\end{align*}
In this appendix, we report the convergence properties of the FE formulations of the linear boundary value problems (5.4)–(5.5) and (5.20)–(5.22) defined in Chapter 5 for the macroscopic elastic dielectric response of dielectric elastomer composites comprising spherical particles and aligned fibers of circular cross section.

### C.1 Convergence study of the FE formulation of the boundary value problems (5.4)–(5.5)

We begin with the convergence properties of the FE formulation of the linear boundary value problems (5.4)–(5.5). For simplicity, we conduct the convergence studies on the general boundary value problems (2.36)–(2.37) in lieu of their periodic formulations (5.4)–(5.5).

For definitiveness, we consider a coated sphere made up of a spherical core with radius $R_p$ (centered for convenience at the origin), surrounded (coated) by a spherical shell of internal radius $R_p$ and external radius $R_m$. This coated sphere is denoted by $\mathcal{CS} = \{ \mathbf{X} : |\mathbf{X}| \leq R_m \}$, while its boundary is denoted as $\partial \mathcal{CS} = \{ \mathbf{X} : |\mathbf{X}| = R_m \}$. Similar to Section 4.1, this choice of geometry allows for non-trivial solutions for boundary value problems (2.36)–(2.37) to be worked out analytically.
C. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

C.1.1 Hybrid FE formulation of the boundary value problem (2.36)

We begin by investigating the convergence properties of the FE hybrid formulation of the linear boundary value problem (2.36). For simplicity, we shall restrict attention here to the vector field $\Gamma_{12}$ corresponding to the components $\Gamma_{i12}$ of the third order tensor $\Gamma$. Consistent with the results presented in Section 6.1, we consider the constituents of the coated sphere to be isotropic, so that the local modulus of elasticity $L$ is given by

$$L = \begin{cases} 
L^{(2)} = 2\mu_p K + 3\kappa_p J & \text{if } |X| \leq R_p \\
L^{(1)} = 2\mu K + 3\kappa J & \text{if } R_p < |X| \leq R_m 
\end{cases}, \quad (C.1)$$

where the fourth-order tensors $K$ and $J$ are given by (3.20). In order to simplify the exposition, we consider the modified boundary value problem for $\Gamma_{i12}$ given by

$$\left[ (\kappa(X) - \frac{2}{3} \mu(X)) \Gamma_{k12,k} \delta_{ij} + 2\mu(X) \Gamma_{(i12,j)} \right]_{,j} = 0, \quad X \in CS$$

with $\Gamma_{i12} = \delta_{i1} X_2 + \delta_{i2} X_1, \quad X \in \partial CS$, \quad (C.2)

where $\mu(X)$ and $\kappa(X)$ correspond to the local shear and bulk moduli

$$\mu(X) = \begin{cases} 
\mu_p & \text{if } |X| \leq R_p \\
\mu & \text{if } R_p < |X| \leq R_m 
\end{cases}, \quad \kappa(X) = \begin{cases} 
\kappa_p & \text{if } |X| \leq R_p \\
\kappa & \text{if } R_p < |X| \leq R_m 
\end{cases}. \quad (C.3)$$

The boundary value problem (C.2) only differs from the original problem (2.36) by the symmetrization with respect to the indices $i, j$ indicated by the subscript $(i12,j)$ and by the symmetrized boundary conditions.

Hybrid formulation. As anticipated in Section 5.1, we consider a hybrid FE formulation of (C.2) to account for the (nearly)-incompressible mechanical behavior of typical elastomers and to avoid subsequent the well-known volumetric locking issue of the classical displacement-based formulation. In this context, a hybrid formulation\(^1\) of (C.2) reads as

$$\begin{cases} 
[2\mu(X) \Gamma_{(i12,j)} + p \delta_{ij},j = 0, \quad X \in CS \\
\Gamma_{k12,k} - \frac{3}{3\kappa(X) - 2\mu(X)} p = 0, \quad X \in \partial CS 
\end{cases}$$

with $\Gamma_{i12} = \delta_{i1} X_2 + \delta_{i2} X_1, \quad X \in \partial CS$. \quad (C.4)

where $p$ is a pressure-like field. With help of solid harmonics (see, e.g., Love, 1906) and in a similar fashion to Section 4.1.1, it is not difficult to obtain analytical expressions for the displacement-like

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\(^1\)Accounting for the (nearly)-incompressible behavior of certain classes or materials like elastomers or fluids can also be achieved by other formulations such as mixed formulations; see, e.g. Chapter 1 in Boffi et al. (2012).
vector field $\Gamma_{12}$ and pressure-like scalar field $p$ solutions of (C.4). They read as

\begin{align*}
\Gamma_{112} &= f(R)X_2 + g(R)X_1^2X_2, \\
\Gamma_{212} &= f(R)X_1 + g(R)X_1X_2^2, \\
\Gamma_{312} &= g(R)X_1X_2X_3, \\
p &= \left[\kappa(X) - \frac{2}{3}\mu(X)\right]\Gamma_{k12,k}. \tag{C.5}
\end{align*}

In the above expressions, $f$ and $g$ are functions of $R = |X|$ given by

\begin{align*}
f &= \begin{cases}
A_1 + A_3R^2 & \text{if } 0 \leq R \leq R_p \\
B_1 + \frac{B_2}{R^5} + B_3R^2 + \frac{B_4}{R^3} & \text{if } R_p < R \leq 1
\end{cases}, \tag{C.6}
\end{align*}

and

\begin{align*}
g &= \begin{cases}
-\frac{2(6\kappa + 17\mu)}{15\kappa + 11\mu}A_3 & \text{if } 0 \leq R \leq R_p \\
-\frac{B_2}{R^7} - \frac{2(6\kappa + 17\mu)}{15\kappa + 11\mu}B_3 + \left[1 + \frac{3\kappa}{\mu}\right]\frac{B_4}{R^5} & \text{if } R_p < R \leq 1
\end{cases}.
\end{align*}

Here, the six constants $A_1, A_3, B_1, B_2, B_3, B_4$ are solutions of a linear system of six equations stemming from the continuity of $\Gamma_{112}$ and $2\mu(X)\Gamma_{i12,j}X_j + pX_i$ across the material interface at $R = R_p$ and boundary conditions $\Gamma_{i12} = \delta_{i1}X_2 + \delta_{i2}X_1$ at $R = R_m$. Due to their bulkiness, we do not report their explicit expressions here.

**Hybrid FE formulation.** The weak formulation of (C.4) is then given by

\begin{align*}
\int_{\mathcal{CS}} \left\{2\mu(X)\Gamma_{i12,j} + p\delta_{ij}\right\}v_{i,j} \, dX &= 0 \quad \forall v \in \mathcal{U}^0, \\
\int_{\mathcal{CS}} \left\{\Gamma_{k12,k} - \frac{3}{3\kappa(X) - 2\mu(X)}p\right\}q \, dX &= 0 \quad \forall q \in \mathcal{Q}, \tag{C.8}
\end{align*}

where $\Gamma_{12} \in \mathcal{U}$, $p \in \mathcal{Q}$, and the functional spaces $\mathcal{U}, \mathcal{U}^0$, and $\mathcal{Q}$ are defined as

\begin{align*}
\mathcal{U} &= \{ u \in H^1(\mathcal{CS}), \ u_i = \delta_{i1}X_2 + \delta_{i2}X_1, \ X \in \partial\mathcal{CS}\}, \\
\mathcal{U}^0 &= \{ u \in H^1(\mathcal{CS}), \ u_i = 0, \ X \in \partial\mathcal{CS}\}, \\
\mathcal{Q} &= \{ q \in L^2(\mathcal{CS})\}.
\end{align*}

In the above definitions, $H^1(\mathcal{CS})$ denotes the Sobolev space of vector fields that are, together with their gradient, square integrable on the coated sphere and $L^2(\mathcal{CS})$ stands for the Hilbert space of square integrable scalar fields on the coated sphere. We denote for later reference by $|| \cdot ||_{L^2}$ the standard norm associated with $L^2(\mathcal{CS})$ and by $|| \cdot ||_{H^1}$ the Sobolev norm associated with $H^1(\mathcal{CS})$. 
C. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

As already mentioned in Section 5.1, we employ the finite-element method to obtain approximate solutions for $\Gamma_{12}$ and $p$ within finite-dimensional subspaces $U^h, U^{0,h}, Q^h$ of the functional spaces (C.9). Since the construction of the finite dimensional subspaces and computation of the approximate solutions is of standard matter, we do not report them here. We only mention as essential details that the coated sphere $CS$ is partitioned with standard isoparametric 10-node tetrahedral finite elements, allowing therefore the construction of a trial field $\Gamma^h_{12}$ that is continuous piecewise-quadratic, and of a trial field $p^h$ that is discontinuous piece-wise-constant.

It is crucial to note here that the choice of FE discretization (or equivalently of finite-dimensional subspaces $U^h, U^{0,h}, Q^h$) must be carried out appropriately in order to ensure stability, convergence and optimality of the FE approximations. Specifically, for (nearly-)incompressible materials, subspaces $U^h, U^{0,h}, Q^h$ must be selected so as to satisfy the so-called inf-sup condition

$$\inf_{q^h \in Q^h \setminus \{0\}} \sup_{v^h \in U^{0,h} \setminus \{0\}} \frac{\int_{CS} q^h v^h_{k,k} \, dX}{\|v^h\|_{H^1} \|q^h\|_{L^2}} = \gamma_h \geq \gamma > 0, \quad (C.10)$$

from which, the optimality relation

$$||\Gamma^h_{12} - \Gamma_{12}||^2_{H^1} + (1 + \alpha) ||p^h - p||^2_{L^2} \leq \inf_{u^h \in U^h} (||\Gamma_{12} - u^h||^2_{H^1} + (1 + \alpha) ||p - p^h||^2_{L^2}) \quad (C.11)$$

and optimal error bounds

$$||\Gamma^h_{12} - \Gamma_{12}||_{H^1} + ||p^h - p||_{L^2} \leq C h (||\Gamma_{12}||_{H^1} + ||p||_{L^2}) \quad (C.12)$$

can be rigourously derived; see, e.g. Chapter 5 in Boffi et al. (2012). In the above expressions,

$$\alpha = \min_{X \in CS} \left\{ \frac{3}{3\kappa(X) - 2\mu(X)} \right\}, \quad (C.13)$$

$\gamma$ and $C$ stands for non-negative constants independent of $\alpha$ and the mesh-size measured by $h$, the maximum element diameter. It can be shown in turn with help of interpolation theory (see, e.g., Chapter 4 in Brenner and Scott (2007)) that, for “nice” enough domains such as the coated sphere $CS$, the simpler optimal error bounds hold

$$||\Gamma^h_{12} - \Gamma_{12}||_{L^2} \approx C_1 h^{k+1} \quad \text{and} \quad ||p^h - p||_{L^2} \approx C_2 h^k, \quad (C.14)$$

where $C_1, C_2$ are non-negative constants independent of $h$ and $k$ is the complete polynomial order used in the space $U^h$. We refer the interested reader to the monograph by Boffi et al. (2012) and

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2 Other conditions on the problem formulation itself are also required for the derivation of these results. Satisfied for the problem at hand, these conditions are here independent of the choice of FE discretization and as such, are not discussed for the sake of brevity.
the review paper by Bathe (2001) for precise discussion of the above results and details on their derivation.

For instance, the above-selected hybrid FE discretization — piecewise-quadratic continuous field $\Gamma_{12}^h$ with piecewise-constant discontinuous field $p^h$ — has been shown not to satisfy the inf-sup condition (C.10) in the limit of incompressible materials (see, e.g. Chapter 8 in Boffi et al. (2012)). It is however well known that FE discretizations that do not satisfy the inf-sup condition (C.10) can yet provide acceptable results (see, e.g., Chapelle and Bathe, 1993; Bathe, 2001; Section 5.6.2 in Boffi et al., 2012). Quoting Fortin (1981) on the mathematically analogous problem of Stokes flow, “Computations were done, with success, using theoretically dubious elements or at best using elements for which theory was silent. On the other hand, elements for which convergence proofs were available were treated with suspicion by code developers.”

In this spirit, we show that our choice of hybrid FE discretization provides acceptable results for the relatively large yet finite values of bulk modulus to shear modulus ratio $\kappa/\mu = 10^3$ employed subsequently in Section 6.1 to approximate the nearly-incompressible behavior of typical elastomers. Convergence studies were conducted for the boundary problem (C.4), with, for definitiveness, $\mu = 1.0$ MPa, $\kappa/\mu = 10^3$, $\mu_p/\mu = 10^3$, $\kappa_p/\mu = 10^3$, $R_p = 0.2$ mm, $R_m = 0.5$ mm. In this context, the evolution with the maximum element diameter $h$ of the normalized $L^2$-norms of (a) the error in the displacement-like vector field $\Gamma_{12}^h - \Gamma_{12}$ and (b) the pressure-like field $p^h - p$ are presented on Fig. C.1. Figure C.1 reveals that the numerical solutions converge with the refinement of the mesh to the exact solutions (C.5) as $||\Gamma_{12}^h - \Gamma_{12}||_{L^2} \approx O(h^{1.9})$ and $||p^h - p||_{L^2} \approx O(h^{0.6})$. For comparison, hybrid FE discretizations satisfying (C.10) with quadratic approximation of the displacement-like field $\Gamma_{12}$ (such as the one described in Section 8.3), lead to optimal convergence as $||\Gamma_{12}^h - \Gamma_{12}||_{L^2} = O(h^{3.0})$ and $||p^h - p||_{L^2} = O(h^{2.0})$.

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Various choices of hybrid FE discretizations for the trial displacement and pressure fields have been shown analytically to or not to satisfy the inf-sup condition (C.10) in two and three spatial dimensions. Numerical tests have also been devised (Chapelle and Bathe, 1993) to estimate $\gamma_h$ in (C.10) and thus investigate the satisfaction of the inf-sup condition (C.10) for choices of hybrid FE discretization for which analytical results could not be established.
C. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

C.1.2 FE formulation of the boundary value problem (2.37)

Next, we investigate the convergence properties of the FE discretization of the boundary value problem (2.37). Again, for consistency with the results presented in Section 6.1, we consider that the core and shell of the coated sphere $\mathcal{CS}$ are made out of isotropic linear dielectric materials so that the local permittivity tensor $\epsilon$ entering in (2.37) takes on the form

$$\epsilon = \begin{cases} 
\epsilon^{(2)} = \varepsilon_p \mathbf{I} & \text{if } |\mathbf{X}| \leq R_p \\
\epsilon^{(1)} = \varepsilon \mathbf{I} & \text{if } R_p < |\mathbf{X}| \leq R_n \end{cases}.$$  \hspace{1cm} (C.15)

In this context, the boundary value problem (2.37) for the vector field $\gamma$ specializes to

$$[\varepsilon(\mathbf{X}) \gamma_{i,j}]_{,i} = 0, \ \mathbf{X} \in \mathcal{CS} \quad \text{with} \quad \gamma_i = \delta_{ij} X_j, \ \mathbf{X} \in \partial \mathcal{CS}. \hspace{1cm} (C.16)$$

where $\varepsilon(\mathbf{X})$ corresponds to the local dielectric permittivity

$$\varepsilon(\mathbf{X}) = \begin{cases} 
\varepsilon_p & \text{if } |\mathbf{X}| \leq R_p \\
\varepsilon & \text{if } R_p < |\mathbf{X}| \leq R_n \end{cases}.$$  \hspace{1cm} (C.17)

The vector field $\gamma$ solution to (C.16) is simply given by

$$\gamma = - \left( a + \frac{b}{R^2} \right) \mathbf{X},$$  \hspace{1cm} (C.18)
where the notation $R \equiv |\mathbf{X}|$ is recalled and where

$$a = \begin{cases} 
\frac{3\varepsilon R^3}{(\varepsilon - \varepsilon_p)R^3_p - (2\varepsilon + \varepsilon_p)R^3_m} & \text{if } 0 \leq R \leq R_p \\
\frac{(2\varepsilon + \varepsilon_p)R^3_m}{(\varepsilon - \varepsilon_p)R^3_p - (2\varepsilon + \varepsilon_p)R^3_m} & \text{if } R_p \leq R \leq 1 
\end{cases}$$

and

$$b = \begin{cases} 
0 & \text{if } 0 \leq R \leq R_p \\
\frac{-(\varepsilon_p - \varepsilon)R^3_p}{(\varepsilon - \varepsilon_p)R^3_p - (2\varepsilon + \varepsilon_p)R^3_m} & \text{if } R_p \leq R \leq 1 
\end{cases}$$

**FE formulation.** The weak formulation of the problem (C.16) is given by

$$\int_{CS} \left[ \varepsilon(X) \gamma_{i,j} \right] v_{i,j} \, dX = 0$$

where $\gamma \in \mathcal{G}$ and the functional spaces $\mathcal{G}$ and $\mathcal{G}^0$ are defined as

$$\mathcal{G} = \left\{ u \in H^1(\partial CS), \ u_i = X_i, \ X \in \partial CS \right\},$$

$$\mathcal{G}^0 = \left\{ u \in H^1(\partial CS), \ u_i = 0, \ X \in \partial CS \right\}.$$  

Again, we employ the finite-element method to construct finite-dimensional subspaces $\mathcal{G}^h$, and $\mathcal{G}^{0,h}$ of the functional spaces (C.22) to obtain a numerical approximation of $\gamma$. For this linear problem, the optimality relation

$$||\gamma^h - \gamma|| \leq \inf_{\mathbf{v}^h \in \mathcal{G}^h} ||\gamma - \mathbf{v}^h||_{H^1}$$

and optimal error bound

$$||\gamma^h - \gamma||_{H^1} \leq C h ||\gamma||_{H^1}$$

can be rigorously derived. It can then be shown that the simpler optimal error bound

$$||\gamma^h - \gamma||_{L^2} \approx C_1 h^{k+1}$$

holds for the coated sphere $CS$. In the above expressions, $C$ and $C_1$ are non-negative constants independent of the maximum mesh size $h$, and $k$ is the complete polynomial order used in the space $\mathcal{G}^h$. We refer the interested reader to Chapter 2 in Brenner and Scott (2007) for complete discussion and derivation of these results.

The convergence study was carried out for the case when the coated sphere $CS$ is partitioned with standard isoparametric 10-node tetrahedral finite elements, allowing therefore the construction
C. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

of a trial field $\gamma^h$ that is continuous and piecewise-quadratic. For definitiveness, the parameters entering in (C.16) were chosen as $\varepsilon = 30.0 \times 10^{-12}$ F/m, $\varepsilon_p = 10^2 \varepsilon$, $R_p = 0.2$ mm, $R_m = 0.5$ mm. In this context, the evolution with the maximum element diameter $h$ of the normalized $L^2$-norm of the error $\gamma^h - \gamma$ is presented on Fig. C.2. As expected, the numerical solutions $\gamma^h$ converges to the exact solution (C.18) as $||\gamma^h - \gamma||_{L^2} \approx O(h^{3.0})$ with the refinement of the mesh.

![Figure C.2: Evolution of the normalized $L^2$-norms of the error $\gamma^h - \gamma$ with the maximum element diameter $h$ for the boundary value problem (C.16), with $\varepsilon = 30.0 \times 10^{-12}$ F/m, $\varepsilon_p = 10^2 \varepsilon$, $R_p = 0.2$ mm, $R_m = 0.5$ mm.](image)

C.2 Convergence study of the FE formulation of the boundary value problems (5.20)–(5.22)

We report then the convergence properties of the FE formulation of the linear boundary value problems (5.20)–(5.22). For simplicity, we conduct the convergence studies on the general boundary value problems (5.15), (5.17), and (5.19) instead of their periodic formulation (5.20)–(5.22). However, because the boundary value problem (5.17) is the mathematical analogous of problem (5.19) for a scalar field, explicit converge results for this boundary value problem are not reported for conciseness.

For definitiveness, we consider a coated circle in the $(X_1, X_2)$ plane (centered for convenience at the origin) made up of a circular core with radius $R_p$, surrounded (coated) by a circular shell of internal radius $R_p$ and external radius $R_m$. This coated circle is denoted by $CC = \{ X : \sqrt{X_1^2 + X_2^2} \leq R_m \}$, while its boundary is denoted as $\partial CC = \{ X : \sqrt{X_1^2 + X_2^2} = R_m \}$. Similar to Section 4.2, this choice of geometry allows for non-trivial solutions for boundary value problems (5.15) and (5.19) to be worked out analytically.
C. Hybrid FE formulation for the effective response of DECs in the small-deformation limit

C.2.1 Hybrid FE discretization of the boundary value problem (5.15)

We begin with by investigating the convergence properties of the hybrid FE discretization of the linear boundary value problem (5.15). For simplicity, we restrict our attention here to the two-dimensional vector field $\Gamma^{12}$ corresponding to the components $\Gamma^{\eta\iota\eta\iota}$ of the third order tensor $\Gamma$. Here and subsequently, Greek indices range from 1 to 2. In order to simplify the exposition, we consider the modified boundary value problem for $\Gamma^{\eta\iota\eta\iota}$ given by

$$\lambda(X_1, X_2) \Gamma_{\nu\iota\nu\iota} + 2\mu(X_1, X_2) \Gamma_{(\eta\iota\iota,\iota)}, \iota = 0, \ X \in CC, \quad (C.26)$$

with $\Gamma_{\eta\iota\eta\iota} = \delta_{\eta\iota} X_2 + \delta_{\eta\iota} X_1$, $X \in \partial CC$,

where $\lambda(X_1, X_2)$ and $\mu(X_1, X_2)$ correspond to the local Lamé parameters

$$\lambda(X_1, X_2) = \begin{cases} \lambda_p & \text{if} \ \sqrt{X_1^2 + X_2^2} \leq R_p \\ \lambda & \text{if} \ R_p < \sqrt{X_1^2 + X_2^2} \leq R_n \end{cases}, \quad \mu(X_1, X_2) = \begin{cases} \mu_p & \text{if} \ \sqrt{X_1^2 + X_2^2} \leq R_p \\ \mu & \text{if} \ R_p < \sqrt{X_1^2 + X_2^2} \leq R_n \end{cases}. \quad (C.27)$$

The boundary value problem (C.26) differs from (5.15) by the symmetrization with respect to the indices $\iota, \eta$ indicated by the subscript $(\eta\iota\iota,\iota)$ and by the symmetrized boundary conditions.

**Hybrid formulation.** As anticipated in Section 5.2, we consider a hybrid FE formulation of (C.26) to account for the (nearly)-incompressible mechanical behavior of typical elastomers and to avoid subsequent the well-known volumetric locking issue of the classical displacement-based formulation. In this context, a hybrid formulation reads as

$$\begin{cases} [2\mu(X_1, X_2) \Gamma_{(\eta\iota\iota,\iota)} + p \delta_{\eta\iota}], \iota = 0 \\ \Gamma_{\iota\iota} - \frac{p}{\lambda(X_1, X_2)} = 0 \end{cases}, \ X \in CC \quad \text{with} \quad \Gamma_{\eta\iota\eta\iota} = \delta_{\eta\iota} X_2 + \delta_{\eta\iota} X_1, \ X \in \partial CC. \quad (C.28)$$

With help of solid harmonics (see, e.g., Bland, 1965) and in a similar fashion to Section 4.2.1, it is not difficult to obtain analytical expressions for the displacement-like vector field $\Gamma_{12}$ and pressure-like scalar field $p$ solutions of (C.28). They read as

$$\begin{align*}
\Gamma_{112} &= f(R) X_2 + g(R) X_1^2 X_2, \\
\Gamma_{212} &= f(R) X_1 + g(R) X_1 X_2^2, \\
p &= \lambda(X) \Gamma_{\eta\iota\eta\iota}. \quad (C.29)
\end{align*}$$
In the above expressions, \( f \) and \( g \) are functions of \( R = \sqrt{X_1^2 + X_2^2} \) given by

\[
f = \begin{cases} 
  A_1 + A_3 R^2 & \text{if } 0 \leq R \leq R_p \\
  B_1 + \frac{B_2}{R^4} + B_3 R^2 + \frac{B_4}{R^2} & \text{if } R_p < R \leq 1
\end{cases}
\] (C.30)

and

\[
g = \begin{cases} 
  \frac{2(\lambda_p + 3\mu_p)}{2\lambda_p + 3\mu_p} A_3 & \text{if } 0 \leq R \leq R_p \\
  -4 \frac{B_2}{R^6} - \frac{2(\lambda + 3\mu)}{2\lambda + 3\mu} B_3 + 2 \left[ 1 + \frac{\lambda}{\mu} \right] \frac{B_4}{R^4} & \text{if } R_p < R \leq 1
\end{cases}
\] (C.31)

In the above expressions, the six constants \( A_1, A_3, B_1, B_2, B_3, B_4 \) are solutions of a linear system of six equations stemming from the continuity of \( \Gamma_{12} \) and \( 2\mu(X_1, X_2)\Gamma_{(12,\eta)}X_\eta + pX_\iota \) across the material interphase at \( R = R_p \) and boundary conditions \( \Gamma_{112} = \delta_{11}X_2 + \delta_{12}X_1 \) at \( R = R_m \). Due to their bulkiness, we do not report their explicit expressions here.

**Hybrid FE formulation.** The weak formulation of (C.28) is then given by

\[
\int_{CC} \left\{ 2\mu(X)\Gamma_{(12,\eta)} + p\delta_\iota \right\} v_{\iota,\eta} dX_1 dX_2 = 0 \quad \forall v \in U^0,
\]

\[
\int_{CC} \left\{ \Gamma_{k12,k} - \lambda(X)p \right\} q dX_1 dX_2 = 0 \quad \forall q \in Q,
\] (C.32)

where \( \Gamma_{12} \in U, \ p \in Q, \) and the functional spaces \( U, U^0, \) and \( Q \) are defined as

\[
U = \left\{ u \in H^1(CC), \ u_\iota = \delta_{11}X_2 + \delta_{12}X_1, \ X \in \partial\mathcal{S} \right\},
\]

\[
U^0 = \left\{ u \in H^1(CC), \ u_\iota = 0, \ X \in \partial CC \right\},
\]

\[
Q = \left\{ q \in L^2(CC) \right\}.
\] (C.33)

Here \( H^1(\mathcal{S}) \) denotes the Sobolev space of two-dimensional vector fields that are, together with there gradient, square integrable on the coated circle, and \( L^2(\mathcal{S}) \) stands for the Hilbert space of square integrable scalar fields on the coated circle.

As indicated in Section 5.2, we employ the finite-element method to construct finite-dimensional subspaces \( U^h, U^{0,h}, Q^h \) of the functional spaces (C.9) so as to obtain numerical approximations for the solutions of (C.28). As essential elements, we only mention that the coated circle \( CC \) is partitioned with standard isoparametric 8-node quadrilateral finite elements, allowing therefore the construction of a continuous and piecewise-biquadratic trial field \( \Gamma_{12}^h \), and of a continuous and piece-wise linear trial field \( p^h \).
Similar to the three-dimensional problem discussed in Section C.1.1, the choice of FE discretization, is critical to ensure the convergence of the numerical solutions. Again, when an inf-sup condition analogous to (C.10) is satisfied, error bounds identical to (C.14) can be derived. However, the present choice of hybrid FE discretization — with a continuous piecewise-biquadratic trial field $\mathbf{G}^h_{12}$ and a discontinuous piecewise-linear trial field $p^h$ — does not satisfy the underlying inf-sup condition.

Yet, we show that this hybrid FE discretization provides acceptable results even for the relatively large values of Lamé parameters ratio $\lambda/\mu = 10^3$ employed in Section 6.2 to model the nearly-incompressible behavior of typical elastomers. Convergence studies were conducted for the boundary problem (C.28), with, for definitiveness, $\mu = 1.0$ MPa, $\lambda/\mu = 10^3$, $\mu_p/\mu = 10^3$, $\lambda_p/\mu = 10^3$, $R_p = 0.2$ mm, $R_m = 0.5$ mm. In this context, the evolution with the maximum element diameter $h$ of the normalized $L^2$-norms of (a) the error in the displacement-like vector field $\mathbf{G}^h_{12} - \mathbf{G}_{12}$ and (b) the pressure-like field $p^h - p$ are presented on Fig. C.3. Figure C.3 reveals that the numerical solutions $\mathbf{G}^h_{12}$ and $p^h$ converge to the exact solutions (C.29) as $||\mathbf{G}^h_{12} - \mathbf{G}_{12}||_{L^2} \approx O(h^{3.0})$ and $||p^h - p||_{L^2} \approx O(h^{1.6})$. For comparison, hybrid FE discretizations satisfying the inf-sup condition with quadratic approximation of the displacement-like field $\mathbf{G}_{12}$, lead to optimal convergence as $||\mathbf{G}^h_{12} - \mathbf{G}_{12}||_{L^2} = O(h^{3.0})$ and $||p^h - p||_{L^2} = O(h^{2.0})$. 

![Graphs of (a) the error in the displacement-like vector field and (b) the pressure-like field, normalized by the $L^2$-norms of the corresponding exact fields, with the maximum element diameter $h$ for the boundary value problem (C.28), with for definitiveness, $\mu = 1.0$ MPa, $\lambda/\mu = 10^3$, $\mu_p/\mu = 10^3$, $\lambda_p/\mu = 10^3$, $R_p = 0.2$ mm, $R_m = 0.5$ mm.](image-url)
Next, we investigate the convergence properties of the FE discretization of the boundary value problem (5.19). (5.19) specializes for the two-dimensional vector field $\gamma$ to

$$[\varepsilon(X_1, X_2)\gamma_{\eta, \iota}, \eta] = 0, \ X \in \mathcal{C}\mathcal{C} \quad \text{with} \quad \gamma_{\eta} = X_{\eta}, \ X \in \partial\mathcal{C}\mathcal{C}. \quad \text{(C.34)}$$

where $\varepsilon(X_1, X_2)$ corresponds to the local dielectric permittivity

$$\varepsilon(X_1, X_2) = \begin{cases} \varepsilon_p & \text{if } \sqrt{X_1^2 + X_2^2} \leq R_p \\ \varepsilon & \text{if } R_p < \sqrt{X_1^2 + X_2^2} \leq R_m \end{cases} \quad \text{(C.35)}$$

The vector field $\gamma$ solution to (C.34) is simply given by

$$\gamma_{\iota} = -\left(a + \frac{b}{R^2}\right)X_{\iota}, \quad \text{(C.36)}$$

where we recall the notation $R = \sqrt{X_1^2 + X_2^2}$ and where

$$a = \begin{cases} \frac{2\varepsilon R_m^2}{(\varepsilon - \varepsilon_p)R_p^2 - (\varepsilon + \varepsilon_p)R_m^2} & \text{if } 0 \leq R \leq R_p \\ \frac{(\varepsilon_p - \varepsilon)R_p^2 - (\varepsilon + \varepsilon_p)R_m^2}{(\varepsilon_p - \varepsilon)R_p^2 - (\varepsilon + \varepsilon_p)R_m^2} & \text{if } R_p < R \leq 1 \end{cases} \quad \text{(C.37)}$$

and

$$b = \begin{cases} 0 & \text{if } 0 \leq R \leq R_p \\ \frac{-(\varepsilon_p - \varepsilon)R_p^2R_m^2}{(\varepsilon_p - \varepsilon)R_p^2 - (\varepsilon + \varepsilon_p)R_m^2} & \text{if } R_p < R \leq 1 \end{cases} \quad \text{(C.38)}$$

**FE formulation.** The weak formulation of the boundary value problem (C.34) reads as

$$\int_{\mathcal{C}\mathcal{C}} [\varepsilon(X)\gamma_{\eta, \iota}] v_{\eta, \iota} \ dX_1 \ dX_2 = 0 \quad \forall v \in \mathcal{G}^0 \quad \text{(C.39)}$$

where $\gamma \in \mathcal{G}$ and the functional spaces $\mathcal{G}$ and $\mathcal{G}^0$ are defined as

$$\mathcal{G} = \{ u \in H^1(\mathcal{C}\mathcal{S}), \ u_{\iota} = X_{\iota}, \ X \in \partial\mathcal{C}\mathcal{S} \},$$

$$\mathcal{G}^0 = \{ u \in H^1(\mathcal{C}\mathcal{S}), \ u_{\iota} = 0, \ X \in \partial\mathcal{C}\mathcal{S} \}. \quad \text{(C.40)}$$

Again, we compute a numerical approximation to the solution of (C.34) via the finite-element method, essentially constructing finite-dimensional subspaces $\mathcal{G}^h$, and $\mathcal{G}^{0,h}$ of the functional spaces (C.40). We only mention that the coated circle $\mathcal{C}\mathcal{C}$ is partitioned with standard isoparametric 8-node quadrilateral finite elements, allowing therefore the construction of a trial field $\gamma^h$ that is continuous and piecewise-biquadratic.
Specifically, convergence studies were conducted for the boundary problem (C.34), with, for definitiveness, the choice of parameters \( \varepsilon = 30.0 \times 10^{-12} \text{ F/m}, \varepsilon_p = 10^2 \varepsilon, R_p = 0.2 \text{ mm}, R_a = 0.5 \text{ mm} \). In this context, the evolution with the maximum element diameter \( h \) of the normalized \( L^2 \)-norms of the error \( \gamma^h - \gamma \) is presented on Fig. C.4. As expected, the numerical solution \( \gamma^h \) converges optimally to the exact solution \( \gamma \) as \( ||\gamma^h - \gamma||_{L^2} \approx O(h^{3.0}) \) with the refinement of the mesh.

![Figure C.4: Evolution of the normalized \( L^2 \)-norms of the error \( \gamma^h - \gamma \) with the maximum element diameter \( h \) for the boundary value problem (C.34), with \( \varepsilon = 30.0 \times 10^{-12} \text{ F/m}, \varepsilon_p = 10^2 \varepsilon, R_p = 0.2 \text{ mm}, R_a = 0.5 \text{ mm} \).]
D

Nonlinear electroelastic deformations of dielectric elastomer composites

This appendix contains supplementary material for Chapters 7 and 8 addressing the macroscopic elastic dielectric response of dielectric elastomer composites under finite deformations and finite electric fields.

D.1 The coefficients $\alpha_{mnpq}$

The fifteen coefficients $\alpha_{mnpq}$ in the Hamilton-Jacobi equation (7.12) read as follows:

$$\alpha_{2000} = \frac{2\Gamma_F \lambda_1^3 \lambda_2^2 \left( \lambda_1^6 - 2\lambda_1^4 + 2\lambda_1 \lambda_2^2 - 1 \right)}{3 \left( \lambda_1^2 - \lambda_2^2 \right)^2 \left( \lambda_1^2 \lambda_2^2 - 1 \right)^2} - \frac{\Gamma_F \lambda_1^5 \lambda_2^3 \left( 3\lambda_1^2 \lambda_2^2 - 2\lambda_1 \lambda_2^2 - 1 \right)}{3 \left( \lambda_1^2 - \lambda_2^2 \right)^2 \left( \lambda_1^2 \lambda_2^2 - 1 \right)^2},$$

$$\alpha_{0200} = \frac{2\Gamma_F \lambda_1^3 \lambda_2^2 \left( \lambda_1^4 - 2\lambda_1^2 + 2\lambda_1 \lambda_2^2 + 1 \right)}{3 \left( \lambda_1^2 - \lambda_2^2 \right)^2 \left( \lambda_1^2 \lambda_2^2 - 1 \right)^2} - \frac{\Gamma_F \lambda_1^5 \lambda_2^3 \left( 3\lambda_1^2 \lambda_2^2 - 2\lambda_1 \lambda_2^2 - 1 \right)}{3 \left( \lambda_1^2 - \lambda_2^2 \right)^2 \left( \lambda_1^2 \lambda_2^2 - 1 \right)^2},$$

$$\alpha_{1100} = \frac{2\Gamma_F \lambda_1^3 \lambda_2^2 \left( \lambda_1^4 + \lambda_1^2 \lambda_2^2 - 2 \right)}{3 \left( \lambda_1^2 - \lambda_2^2 \right)^2 \left( \lambda_1^2 \lambda_2^2 - 1 \right)^2} + \frac{2\Gamma_F \lambda_1^5 \lambda_2^3 \left( 3\lambda_1^2 \lambda_2^2 - 2\lambda_1 \lambda_2^2 - 1 \right)}{3 \left( \lambda_1^2 - \lambda_2^2 \right)^2 \left( \lambda_1^2 \lambda_2^2 - 1 \right)^2} - \frac{2\lambda_1 \lambda_2 \left( \lambda_1^4 + \lambda_2^4 \right)}{3 \left( \lambda_1^2 - \lambda_2^2 \right)^2},$$

(D.1)
\[
\alpha_{00200} = \frac{\Gamma_F \lambda_1 \lambda_2}{(\lambda_1^2 - \lambda_2^2) (\lambda_1^2 \lambda_2 - 1)} \left[ \frac{2 \mathcal{E}_1^2 (\lambda_1^4 \lambda_2^2 - 2 \lambda_1^4 \lambda_2^2 + 2 \lambda_1^2 \lambda_2^4 - 1)}{3 \lambda_1^2 (\lambda_1^2 - \lambda_2^2) (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} \right. \\
+ \frac{\mathcal{E}_2^2 (\lambda_1^2 \lambda_2^2 - 2 \lambda_1^2 \lambda_2^2 + 2 \lambda_1^2 \lambda_2^4 - 1)}{3 \lambda_1^2 (\lambda_1^2 - \lambda_2^2) (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} \left( \lambda_1^2 \lambda_2 - 2 \lambda_1^2 \lambda_2^2 + 1 \right) - \frac{\mu}{\varepsilon} \right] + \\
\frac{\Gamma_F \lambda_1^2 \lambda_2^2}{\lambda_1^2 \lambda_2^2 - 1} \left[ \frac{\mathcal{E}_1^2 (2 \lambda_1^2 \lambda_2^2 - 3 \lambda_1^2 \lambda_2^2 + 1)}{3 \lambda_1^2 (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} + \frac{\mathcal{E}_2^2 (\lambda_1^2 \lambda_2^2 - 3 \lambda_1^2 \lambda_2^2 + 1)}{3 \lambda_1^2 (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} \right. \\
+ \frac{\mathcal{E}_3 \lambda_1^2 \lambda_2^2 (\lambda_1^2 \lambda_2^2 - 2 \lambda_1^2 \lambda_2^2 - 2 \lambda_1^2 \lambda_2^4 - 1)}{3 \lambda_1^2 (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} + \frac{\mu}{\varepsilon} \right] + \\
\frac{\Gamma_F \lambda_1^2 \lambda_2^2}{\lambda_1^2 \lambda_2^2 - 1} \left[ \frac{\mathcal{E}_1^2 (2 \lambda_1^2 \lambda_2^2 - 3 \lambda_1^2 \lambda_2^2 + 1)}{3 \lambda_1^2 (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} + \frac{\mathcal{E}_2^2 (\lambda_1^2 \lambda_2^2 - 3 \lambda_1^2 \lambda_2^2 + 1)}{3 \lambda_1^2 (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} \right. \\
+ \frac{\mathcal{E}_3 \lambda_1^2 \lambda_2^2 (\lambda_1^2 \lambda_2^2 - 2 \lambda_1^2 \lambda_2^2 - 2 \lambda_1^2 \lambda_2^4 - 1)}{3 \lambda_1^2 (\lambda_1^2 \lambda_2 - 1) (\lambda_1^2 \lambda_2 - 1)} + \frac{\mu}{\varepsilon} \right].
\]
\[
\alpha_{00002} = \frac{\Gamma_E \alpha \lambda_1 \lambda_2}{\lambda_1 \lambda_2 - 1} \left[ \frac{E_2 (\lambda_1^2 \lambda_2^2 + 1)^2 (\lambda_1^2 \lambda_2^2 - 2\lambda_1^2 \lambda_2^4 + 1)}{3 \lambda_1^2 (\lambda_1^2 - \lambda_2^2) (\lambda_1^2 \lambda_2^4 - 1)^3} - \frac{2E_3^2 \lambda_1^4 \lambda_2^4 (\lambda_1^2 \lambda_2^2 + \lambda_1^2 \lambda_2^4 - 2)}{3 (\lambda_1^2 \lambda_2^4 - 1)(\lambda_1^2 \lambda_2^4 - 1)} + \frac{\Gamma_E \alpha \lambda_1 \lambda_2}{\lambda_1 \lambda_2 - 1} \right]
\]

\[
\alpha_{00110} = -\frac{8E_1 E_2}{3 (\lambda_1^2 - \lambda_2^2)^3} \left[ \frac{\Gamma_E \lambda_1 \lambda_2}{(\lambda_1^2 - \lambda_2^2)(\lambda_1^2 \lambda_2^4 - 1)} + \frac{\Gamma_E \lambda_1 \lambda_2}{\lambda_1 \lambda_2 - 1} \right] \left[ \frac{(\lambda_1^2 + \lambda_2^2)(\lambda_1^4 - 6\lambda_1^2 \lambda_2^4 + \lambda_2^4)}{4 (\lambda_1^2 - \lambda_2^2)} \right],
\]

\[
\alpha_{00011} = \frac{8E_2 E_3 \lambda_1 \lambda_2^4}{3 (\lambda_1^2 \lambda_2^4 - 1)^3} \left[ \frac{\Gamma_E \lambda_1 \lambda_2}{(\lambda_1^2 - \lambda_2^2)(\lambda_1^2 \lambda_2^4 - 1)} - \frac{\Gamma_E \lambda_1 \lambda_2}{\lambda_1 \lambda_2 - 1} \left[ \frac{\lambda_1 \lambda_2^4 - 2\lambda_1 \lambda_2^4 - 1}{4 (\lambda_1^2 - \lambda_2^2)} \right] \right],
\]

\[
\alpha_{00101} = \frac{8E_1 E_3 \lambda_1 \lambda_2^4}{3 (\lambda_1^2 \lambda_2^4 - 1)^3} \left[ \frac{\Gamma_E \lambda_1 \lambda_2}{(\lambda_1^2 - \lambda_2^2)(\lambda_1^2 \lambda_2^4 - 1)} - \frac{2\Gamma_E \lambda_1 \lambda_2}{\lambda_1 \lambda_2 - 1} \left[ \frac{\lambda_1 \lambda_2^4 - 2\lambda_1 \lambda_2^4 - 1}{4 (\lambda_1^2 - \lambda_2^2)} \right] \right],
\]

\[
\alpha_{10100} = \frac{2E_1}{\lambda_1} \alpha_{20000}, \quad \alpha_{01010} = \frac{E_2}{\lambda_2} \alpha_{11000}, \quad \alpha_{10001} = -\frac{2E_3}{\lambda_1} \alpha_{20000} - \frac{E_3}{\lambda_2} \alpha_{11000}.
\]

\[
\alpha_{01100} = \frac{E_1}{\lambda_1} \alpha_{11000}, \quad \alpha_{01010} = \frac{2E_2}{\lambda_2} \alpha_{02000}, \quad \alpha_{01001} = \frac{2E_3}{\lambda_2} \alpha_{02000} - \frac{E_3}{\lambda_1} \alpha_{11000}.
\]
In the above expressions,
\[
\begin{align*}
\Gamma_F &= \frac{1}{\sqrt{1 - \lambda_1^2}} \mathcal{E}_F \left\{ \frac{\sqrt{\lambda_1^2 \lambda_2^2 - 1}}{2 \sqrt{1 - \lambda_1^2 \lambda_2^2}} \ln\left[ 2 \lambda_1 \lambda_2^2 \left( \sqrt{\lambda_1^2 \lambda_2^4 - 1} + \lambda_1 \lambda_2^2 \right) - 1 \right] ; \frac{\lambda_1^2 \lambda_2^2 - 1}{\lambda_1 \lambda_2^2 - 1} \right\}, \\
\Gamma_E &= \frac{1}{\sqrt{1 - \lambda_1^2}} \mathcal{E}_E \left\{ \frac{\sqrt{\lambda_1^2 \lambda_2^2 - 1}}{2 \sqrt{1 - \lambda_1^2 \lambda_2^2}} \ln\left[ 2 \lambda_1 \lambda_2^2 \left( \sqrt{\lambda_1^2 \lambda_2^4 - 1} + \lambda_1 \lambda_2^2 \right) - 1 \right] ; \frac{\lambda_1^2 \lambda_2^2 - 1}{\lambda_1 \lambda_2^2 - 1} \right\},
\end{align*}
\]
where the functions \( \mathcal{E}_F \) and \( \mathcal{E}_E \), stand for, respectively, the incomplete elliptic integrals of first and second kind:
\[
\mathcal{E}_F \{ \phi; \nu \} = \int_0^\phi [1 - \nu \sin^2 \theta]^{-1/2} \, d\theta, \quad \mathcal{E}_E \{ \phi; \nu \} = \int_0^\phi [1 - \nu \sin^2 \theta]^{1/2} \, d\theta.
\]
(D.3)

**D.2 Asymptotic analysis of the Hamilton-Jacobi equation (7.12)–(7.13) in the limit of infinitely large deformations**

This appendix outlines the derivation of the asymptotic solution (7.17) for the effective free-energy function (7.11) with (7.12)–(7.13) in the limit of infinitely large deformations when \( \lambda_1 \to 0, +\infty \) and/or \( \lambda_2 \to 0, +\infty \).

Numerical solutions of the HJ equation (7.12)–(7.13) for \( \overline{U} = \overline{U}(\lambda_1, \lambda_2, E_1, E_2, E_3, c) \) suggest that
\[
\overline{U}(\lambda_1, \lambda_2, E_1, E_2, E_3, c) = \frac{2f(c) - \mu}{4\mu} \left[ \frac{\lambda_2^2}{\lambda_1 + \lambda_2} + \frac{1}{\lambda_1 \lambda_2} \right] + \frac{2g(c) + \varepsilon}{4\mu} \left[ \frac{E_1^2}{\lambda_1} + \frac{E_2^2}{\lambda_2} + \frac{\lambda_1^2 \lambda_2^2 E_3^2}{\lambda_1 \lambda_2} \right] + \text{HOT}
\]
and hence that
\[
\overline{W}(\lambda_1, \lambda_2, E_1, E_2, E_3, c) = f(c) \left[ \frac{\lambda_2^2}{\lambda_1 + \lambda_2} + \frac{1}{\lambda_1 \lambda_2} \right] + g(c) \left[ \frac{E_1^2}{\lambda_1} + \frac{E_2^2}{\lambda_2} + \frac{\lambda_1^2 \lambda_2^2 E_3^2}{\lambda_1 \lambda_2} \right] + \text{HOT}
\]
(D.5)
for large and small values of \( \lambda_1 \) and/or \( \lambda_2 \), where \( f \) and \( g \) are functions of the volume fraction of particles \( c \). Substituting the ansatz (D.4) in the HJ equation (7.12)–(7.13) and subsequently taking the limit of infinitely large deformations yields a hierarchical system of ordinary differential equations for the functions \( f \) and \( g \). Solving the odes corresponding to the leading order term in the asymptotic expansion renders the asymptotic solutions for \( \overline{W} \) presented next.

In the limit as \( \lambda_1 \to +\infty \), \( \overline{W} \) takes on the form
\[
\overline{W}(\lambda_1, \lambda_2, E_1, E_2, E_3, c) = \frac{2(1 - c)\mu + (1 + 2c)\mu_p}{2[(2 + c)\mu + 2(1 - c)\mu_p]} \frac{\mu \lambda_1^2}{2\varepsilon + (1 - c)\varepsilon_p} \lambda_1 \lambda_2 E_3 + o(\lambda_1^2).
\]
(D.6)
In the opposite limit when \( \lambda_1 \to 0 \), \( \mathcal{W} \) reads as
\[
\mathcal{W}(\lambda_1, \lambda_2, E_1, E_2, E_3) = 2(1-c)\mu + (1 + 2c)\mu_p \mu_1 \frac{1}{2} \lambda_2^2 + \frac{\varepsilon_p}{2[\varepsilon_\lambda + (1-c)\varepsilon_p]} \lambda_1^2 + o(\lambda_1^{-2}). \tag{D.7}
\]
If follows from the symmetry condition \( \mathcal{W}(\lambda_1, \lambda_2, E_1, E_2, E_3) = \mathcal{W}(\lambda_2, \lambda_1, E_2, E_1, E_3) \) that \( \mathcal{W} \) is given by
\[
\mathcal{W}(\lambda_1, \lambda_2, E_1, E_2, E_3, c) = 2(1-c)\mu + (1 + 2c)\mu_p \mu_1 \frac{1}{2} \lambda_2^2 - \frac{\varepsilon_p}{2[\varepsilon_\lambda + (1-c)\varepsilon_p]} \varepsilon_1 \lambda_2^2 E_3^2 + o(\lambda_1^{-2}), \tag{D.8}
\]
when \( \lambda_2 \to +\infty \) and by
\[
\mathcal{W}(\lambda_1, \lambda_2, E_1, E_2, E_3, c) = 2(1-c)\mu + (1 + 2c)\mu_p \mu_1 \frac{1}{2} \lambda_2^2 - \frac{\varepsilon_p}{2[\varepsilon_\lambda + (1-c)\varepsilon_p]} \varepsilon_1 \lambda_2^2 E_3^2 + o(\lambda_1^{-2}), \tag{D.9}
\]
when \( \lambda_2 \to 0 \). Combining (D.6)–(D.9) leads to the result (7.17) presented in the main body of the text.

### D.3 Numerical viscosity solution of the Hamilton-Jacobi equation (7.12)–(7.13)

In this appendix, we outline the main features of the WENO finite-difference scheme employed to compute numerically the viscosity solution of the HJ equation (7.12)–(7.13) for the function \( \overline{U} \) in the effective free energy (7.11); the derivation and complete description of the scheme will be reported elsewhere. For simplicity of exposition, we present the scheme in the context of axisymmetric electromechanical loading conditions with

\[
\lambda_1 = \lambda_2, \quad E_1 = E_2 = 0 \quad \text{and employ the notation} \quad \lambda_1 = x, \quad E_3 = y. \tag{D.10}
\]

This allows us to lay out all essential components of the method in the notationally more amenable context of a HJ equation that involves only two space variables: \( x \in (0, +\infty) \) and \( y \in (-\infty, +\infty) \). The generalization to the HJ equation (7.12)–(7.13) involving all five space variables \( \lambda_1, \lambda_2, E_1, E_2, E_3 \) shall be apparent.

We begin by defining the function \( u(x, y, c) = \overline{U}(x, x, 0, 0, y, c) \) and note that
\[
\frac{\partial \overline{U}}{\partial x}(x, x, 0, 0, y, c) = \frac{1}{2} \frac{\partial u}{\partial x}(x, y, c). \tag{D.11}
\]
D. Nonlinear electroelastic deformations of DECs

Under the axisymmetric conditions of loading (D.10), it follows that the HJ equation (7.12)–(7.13) for \( U \) reduces to the simpler HJ equation for \( u \):

\[
\frac{\partial u}{\partial \tau} + H\left(x, y, c, u, \frac{\partial u}{\partial x}, \frac{\partial u}{\partial y}\right) = 0, \quad u(x, y, 1) = \frac{1}{3} \left(\frac{\mu_2}{\mu} - 1\right) \left[\frac{2x^2 + 1}{x^4} - 3\right] + \frac{\varepsilon - \varepsilon p}{4\mu} x^4 y^2, \quad (D.12)
\]

where the Hamiltonian \( H \) in the pde (D.12) is given by

\[
H(x, y, c, u, p, q) = -\frac{u}{c} - \frac{\beta_1(x)}{c} p^2 - \left(\frac{4y^2 \beta_1(x)}{cx^2} + \frac{\mu \beta_2(x)}{\varepsilon cx^4}\right) q^2 + \frac{4y \beta_1(x)}{cx} pq \quad (D.13)
\]

with

\[
\beta_1(x) = \frac{(2x^6 + 1)x^6}{12(x^6 - 1)^2} - \frac{x^{12} \ln \left[\sqrt{x^{-6} - 1 + x^{-3}}\right]}{4(1 - x^6)^{5/2}} \quad \text{and} \quad \beta_2(x) = x^6 - \frac{\ln \left[\sqrt{x^{-6} - 1 + x^{-3}}\right]}{(x^6 - 1)^{3/2} x^3}. \quad (D.14)
\]

D.3.1 Monotone numerical Hamiltonian

We seek to compute the viscosity solution of (D.12) with Hamiltonian given by (D.13). In their celebrated contribution, Crandall and Lions (1984) proved that first-order monotone schemes are convergent to the viscosity solution (Crandall and Lions, 1983). Considering the space \((x, y)\) to be discretized by a grid with uniform spacing \(h_x\) in the \(x\)-direction and \(h_y\) in the \(y\)-direction, denoting by \(u_{i,j}\) the numerical approximation to the viscosity solution of (D.12) at the space point \((x, y) = (x_i, y_j)\) and time \(c\), that is, \(u(x_i, y_j, c) = u(ih_x, jh_y, c)\), and making use of the notation \(u_{x,i,j}^+ = \Delta_x^+ u_{i,j}/h_x = (u_{i+1,j} - u_{i,j})/h_x\), \(u_{x,i,j}^- = \Delta_x^- u_{i,j}/h_x = (u_{i,j} - u_{i-1,j})/h_x\), \(u_{y,i,j}^+ = \Delta_y^+ u_{i,j}/h_y = (u_{i,j+1} - u_{i,j})/h_y\), \(u_{y,i,j}^- = \Delta_y^- u_{i,j}/h_y = (u_{i,j} - u_{i,j-1})/h_y\), first-order monotone schemes refer to schemes of the form

\[
\frac{d}{dc} u_{i,j} = -\hat{H}\left(x_i, y_j, c, u_{i,j}, u_{x,i,j}^+, u_{x,i,j}^-, u_{y,i,j}^+, u_{y,i,j}^-, u_{x,i,j}, u_{y,i,j}\right), \quad (D.15)
\]

where \(\hat{H}\), the so-called numerical Hamiltonian (also termed flux), is a Lipschitz continuous function such that it is consistent with the Hamiltonian \(H\) in the sense that

\[
\hat{H}(x, y, c, u, p, p, q, q) = H(x, y, c, u, p, q), \quad (D.16)
\]

and it is monotone in the sense that it is nonincreasing in its fifth and seventh arguments and nondecreasing in its sixth and eighth arguments, symbolically,

\[
\hat{H}(x, y, c, u, ↓, ↑, ↓, ↑). \quad (D.17)
\]

There are a number of monotone numerical Hamiltonians that have been proposed in the literature. In this appendix, we make use of the so-called Roe flux with LLF entropy correction (Osher and...
Omitting the dependence on $x, y, c,$ and $u$ to ease notation, the Roe flux with LLF entropy correction reads as:

\[
\hat{H}(p^+, p^-, q^+, q^-) = \begin{cases} 
\mathcal{H}(p^*, q^*) & \text{if } \mathcal{H}_1(p, q) \text{ and } \mathcal{H}_2(p, q) \text{ do not} \\
\mathcal{H}\left(\frac{p^++p^-}{2}, q^*\right) - \nu_1(p^+, p^-) \frac{p^+ - p^-}{2} & \text{change signs in } p \in I(p^-, p^+), \\
\mathcal{H}\left(p^*, \frac{q^++q^-}{2}\right) - \nu_2(q^+, q^-) \frac{q^+ - q^-}{2} & q \in I(q^-, q^+); \\
\hat{H}_{\text{LLF}}(p^+, p^-, q^+, q^-) & \text{otherwise and if } \mathcal{H}_2(p, q) \text{ does} \\
& \text{not change sign in } A \leq p \leq B, \\
& q \in I(q^-, q^+); \\
& \text{otherwise and if } \mathcal{H}_1(p, q) \text{ does} \\
& \text{not change sign in } p \in I(p^-, p^+), \\
& C \leq q \leq D; \\
& \text{otherwise} \\
\end{cases}
\tag{D.18}
\]

where $p^*$ and $q^*$ are defined by

\[
p^* = \begin{cases} 
p^+ & \text{if } \mathcal{H}_1(p, q) \leq 0 \\
p^- & \text{if } \mathcal{H}_1(p, q) \geq 0
\end{cases}, \quad q^* = \begin{cases} 
q^+ & \text{if } \mathcal{H}_2(p, q) \leq 0 \\
q^- & \text{if } \mathcal{H}_2(p, q) \geq 0
\end{cases}.
\tag{D.19}
\]

\[\nu_1 \text{ and } \nu_2 \text{ are defined by}
\]

\[
\nu_1(p^+, p^-) = \max_{p \in I(p^-, p^+), C \leq q \leq D} |\mathcal{H}_1(p, q)|, \quad \nu_2(q^+, q^-) = \max_{q \in I(q^-, q^+), A \leq p \leq B} |\mathcal{H}_2(p, q)|,
\tag{D.20}
\]

and

\[
\hat{H}_{\text{LLF}}(p^+, p^-, q^+, q^-) = \mathcal{H}\left(\frac{p^++p^-}{2}, \frac{q^++q^-}{2}\right) - \nu_1(p^+, p^-) \frac{p^+ - p^-}{2} - \nu_2(q^+, q^-) \frac{q^+ - q^-}{2}.
\tag{D.21}
\]

Here, $\mathcal{H}_1 = \partial \mathcal{H}(p, q) / \partial p, \mathcal{H}_2 = \partial \mathcal{H}(p, q) / \partial q, [A, B] ([C, D])$ denotes the range of values taken by $p^\pm (q^\pm)$ over the entire space $(x, y)$ considered, and $I(a, b) = [\min(a, b), \max(a, b)]$.

As initiated by Osher and Sethian (1988), a formal — yet proven robust over time — approach to compute viscosity solutions with more than first-order accuracy is to still use monotone numerical Hamiltonians $\mathcal{H}$, but now using high-order approximations for the partial derivatives of the function $u$ in place of the first-order finite differences $u_{x,i,j}^\pm = \Delta_x^\pm u_{i,j}/h_x, u_{y,i,j}^\pm = \Delta_y^\pm u_{i,j}/h_y$. We follow this same approach here. Specifically, as described next, we use fifth-order WENO approximations in place of the first-order finite differences $\Delta_x^\pm u_{i,j}/h_x, \Delta_y^\pm u_{i,j}/h_y$.
Before proceeding with the technical details, we note that WENO finite-difference schemes were originally introduced in the 1990s by Jiang and Shu (1996), as a generalization of the pioneering work of Liu et al. (1994) on WENO finite-volume schemes, within the context of hyperbolic conservation laws and have become increasingly popular over the last twenty years as a method of choice to solve numerically convection dominated pdes. The defining feature of WENO schemes is that they provide the means to reach arbitrarily high order accuracy (at least formally) in smooth regions of the solution while being able to describe in a non-oscillatory manner regions of discontinuities or steep gradients. For more details about WENO schemes, including an overview of their increasing application to an admittedly broad range of physical problems, we refer the interested reader to the review of Shu (2009).

**D.3.2 The WENO “space” discretization**

We find it sufficient to restrict attention to discrete space domains of computation defined by regular Cartesian grids of the form \(\{(x_i, y_j) : x_0 < x_1 < ... < x_{m-1} < x_m, \ y_0 < y_1 < ... < y_{n-1} < y_n\}\) with \(x_{i+1} - x_i = h_x = h, \ y_{j+1} - y_j = h_y = h \ \forall (i,j) \in \{0, 1, 2, ..., m\} \times \{0, 1, 2, ..., n\}\), where \(h\) is a prescribed constant. In this setting, for grid points \((x_i, y_j)\) with \(3 \leq i \leq m-2\), the fifth-order WENO approximation of the discrete partial derivative \(u^{-+}_{x,i,j}\) that we utilize in the monotone Hamiltonian (D.18) is given by the formula

\[
u_{x,i,j} = \frac{1}{12} \left( -\frac{\Delta^+_x u_{i-2,j}}{h} + 7\frac{\Delta^+_x u_{i-1,j}}{h} + 7\frac{\Delta^+_x u_{i+1,j}}{h} - \frac{\Delta^+_x u_{i+2,j}}{h} \right) -
\]

\[
g \left( \frac{\Delta^+_x \Delta^+_x u_{i-2,j}}{h^2}, \frac{\Delta^+_x \Delta^+_x u_{i-1,j}}{h^2}, \frac{\Delta^-_x \Delta^+_x u_{i-1,j}}{h^2}, \frac{\Delta^-_x \Delta^+_x u_{i+1,j}}{h^2} \right). \tag{D.22}\]

For grid points \((x_i, y_j)\) with \(2 \leq i \leq m-3\), on the other hand, the fifth-order WENO approximation of \(u^{+-}_{x,i,j}\) is given by

\[
u_{x,i,j} = \frac{1}{12} \left( -\frac{\Delta^+_x u_{i-2,j}}{h} + 7\frac{\Delta^+_x u_{i-1,j}}{h} + 7\frac{\Delta^+_x u_{i+1,j}}{h} - \frac{\Delta^+_x u_{i+2,j}}{h} \right) +
\]

\[
g \left( \frac{\Delta^-_x \Delta^+_x u_{i-2,j}}{h^2}, \frac{\Delta^-_x \Delta^+_x u_{i-1,j}}{h^2}, \frac{\Delta^-_x \Delta^-_x u_{i+1,j}}{h^2}, \frac{\Delta^-_x \Delta^-_x u_{i+1,j}}{h^2} \right). \tag{D.23}\]

Similarly, for grid points \((x_i, y_j)\) with \(3 \leq j \leq n-2\), the WENO approximation of the discrete partial derivative \(u^{-+}_{y,i,j}\) is given from symmetry considerations by the formula

\[
u_{y,i,j} = \frac{1}{12} \left( -\frac{\Delta^+_y u_{i-2,j}}{h} + 7\frac{\Delta^+_y u_{i-1,j}}{h} + 7\frac{\Delta^+_y u_{i+1,j}}{h} - \frac{\Delta^+_y u_{i+2,j}}{h} \right) -
\]

\[
g \left( \frac{\Delta^-_y \Delta^+_y u_{i-2,j}}{h^2}, \frac{\Delta^-_y \Delta^+_y u_{i-1,j}}{h^2}, \frac{\Delta^-_y \Delta^-_y u_{i+1,j}}{h^2}, \frac{\Delta^-_y \Delta^-_y u_{i+1,j}}{h^2} \right). \tag{D.24}\]
For grid points \((x_i, y_j)\) with \(2 \leq j \leq n - 3\), the fifth-order WENO approximation of \(u_{y,i,j}^+\) that we employ is given by

\[
u_{y,i,j}^+ = \frac{1}{12} \left( -\frac{\Delta_y^+ u_{i,j-2}}{h} + 7 \frac{\Delta_y^+ u_{i,j-1}}{h} + \frac{7 \Delta_y^+ u_{i,j}}{h} - \frac{\Delta_y^+ u_{i,j+1}}{h} \right) + g \left( \frac{\Delta_y^+ \Delta_y^+ u_{i,j+2}}{h}, \frac{\Delta_y^- \Delta_y^+ u_{i,j+1}}{h}, \frac{\Delta_y^+ \Delta_y^- u_{i,j}}{h}, \frac{\Delta_y^- u_{i,j-1}}{h} \right).
\]

(D.25)

In expressions (D.22) through (D.25),

\[
g(z_1, z_2, z_3, z_4) = \frac{1}{3} \tilde{\omega}^{(1)} (z_1 - 2z_2 + z_3) + \frac{1}{6} (\tilde{\omega}^{(3)} - \frac{1}{2}) (z_2 - 2z_3 + z_4)
\]

(D.26)

with

\[
\tilde{\omega}^{(l)} = \frac{\gamma^{(l)}}{\sum_{k=1}^{3} (\varepsilon + h^2 \tilde{S}^{(l)})^2}, \quad \gamma^{(1)} = \frac{1}{10}, \quad \gamma^{(2)} = \frac{3}{5}, \quad \gamma^{(3)} = \frac{3}{10}, \quad \epsilon = 10^{-6},
\]

(D.27)

and

\[
\tilde{S}^{(k)} = \frac{13}{12} (z_k - z_{k+1})^2 + [(k - 2)z_k - (k - 3)z_{k+1})^2,
\]

(D.28)

where we have made use again of the standard notation \(\Delta_x^+ v_{k,l} = v_{k+1,l} - v_{k,l}, \Delta_x^- v_{k,l} = v_{k,l} - v_{k-1,l}\), \(\Delta_y^+ v_{k,l} = v_{k,l+1} - v_{k,l}, \Delta_y^- v_{k,l} = v_{k,l} - v_{k,l-1}\).

As specified above, expressions (D.22)–(D.25) are only valid at grid points \((x_i, y_j)\) sufficiently away from the boundaries \(x = x_0, x_m\) and \(y = y_0, y_n\) of the domain of computation. In the regions of the grid where they are not valid, we utilize expressions of similar nature recently put forth which maintain the fifth-order accuracy of the scheme.

**D.3.3 The “time” discretization**

The next and final step in the construction of our scheme is to carry out the discretization of the semi-discrete HJ equation (D.15) in the time variable \(c\). To this end, we employ the fifth-order explicit Runge-Kutta discretization with extended region of stability due to Lawson (1966).

Similar to the space discretization, we discretize time by means of a grid \(c^n < c^{n-1} < ... < c^2 < c^1 < c^0\) with \(c^{k+1} - c^k = \Delta c\) for all \(k = \{0, 1, 2, ..., n\}\), where \(\Delta c\) is a constant. For grid sizes \(h_x = h_y = h\) and \(\Delta c\), we denote by \(u_{i,j}^n\) a numerical approximation to the viscosity solution of (D.15) at the space point \((x, y) = (x_i, y_j)\) and time \(c = c^n\), namely, \(u(x_i, y_j, c^n) = u(x_0 + ih, y_0 + jh, c^0 + n\Delta c)\). Given
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\[ L_{i,j}^{(l)} \{ c \} = -\tilde{H}(x_i, y_j, \mathbf{c}, u_{i,j}^{(l)}, u_{x,i,j}^{(l)+}, u_{x,i,j}^{(l)-}, u_{y,i,j}^{(l)+}, u_{y,i,j}^{(l)-}), \]  

(D.29)

where, again, the numerical Hamiltonian \( \tilde{H} \) is given by the Roe flux (D.18) and \( u_{x,i,j}^{(l)+}, u_{x,i,j}^{(l)-}, u_{y,i,j}^{(l)+}, u_{y,i,j}^{(l)-} \) stand for the fifth-order WENO approximations (D.22)–(D.25) and corresponding approximations applicable on the boundary of the grid, we obtain \( u_{i,j}^{n+1} \) from \( u_{i,j}^{n} \) by following the fifth-order Runge-Kutta procedure:

\[
\begin{align*}
u_{i,j}^{(1)} &= u_{i,j}^{n}, & k_{i,j}^{(1)} &= L_{i,j}^{(1)} \{ c^{n} \}, \\
u_{i,j}^{(2)} &= u_{i,j}^{(1)} + \frac{1}{2}\Delta c k_{i,j}^{(1)}, & k_{i,j}^{(2)} &= L_{i,j}^{(2)} \left\{ c^{n} + \frac{1}{2}\Delta c \right\}, \\
u_{i,j}^{(3)} &= u_{i,j}^{(1)} + \frac{1}{16}\Delta c (3k_{i,j}^{(1)} + k_{i,j}^{(2)}), & k_{i,j}^{(3)} &= L_{i,j}^{(3)} \left\{ c^{n} + \frac{1}{4}\Delta c \right\}, \\
u_{i,j}^{(4)} &= u_{i,j}^{(1)} + \frac{1}{2}\Delta c k_{i,j}^{(3)}, & k_{i,j}^{(4)} &= L_{i,j}^{(4)} \left\{ c^{n} + \frac{1}{2}\Delta c \right\}, \\
u_{i,j}^{(5)} &= u_{i,j}^{(1)} + \frac{3}{16}\Delta c (-k_{i,j}^{(2)} + 2k_{i,j}^{(3)} + 3k_{i,j}^{(4)}), & k_{i,j}^{(5)} &= L_{i,j}^{(5)} \left\{ c^{n} + \frac{3}{4}\Delta c \right\}, \\
u_{i,j}^{(6)} &= u_{i,j}^{(1)} + \frac{1}{7}\Delta c (k_{i,j}^{(1)} + 4k_{i,j}^{(2)} + 6k_{i,j}^{(3)} - 12k_{i,j}^{(4)} + 8k_{i,j}^{(5)}), & k_{i,j}^{(6)} &= L_{i,j}^{(6)} \{ c^{n} + \Delta c \}, \\
u_{i,j}^{n+1} &= u_{i,j}^{n} + \frac{\Delta c}{90} \left( 7k_{i,j}^{(1)} + 32k_{i,j}^{(3)} + 12k_{i,j}^{(4)} + 32k_{i,j}^{(5)} + 7k_{i,j}^{(6)} \right). & 
\end{align*}
\]

(D.30)

The corresponding formulae for the general HJ equation (7.12)–(7.13) involving all five space variables \( \lambda_1, \lambda_2, E_1, E_2, E_3 \) should be now apparent.

D.4 A conforming FE approximation based on Crouzeix-Raviart-type elements

In this appendix, we provide details of the conforming FE method that we employ to construct numerical solutions for the Euler–Lagrange equation (8.107).

D.4.1 The formulation

Seeking the ability to discretize complex domains, we consider partitions of the unit cell that comprise \( N_e \) non-overlapping “curved” tetrahedral elements: \( Y = \bigcup_{e=1}^{N_e} \mathcal{E}^{(e)} \) with \( \mathcal{E}^{(i)} \cap \mathcal{E}^{(j)} = \emptyset \forall i \neq j \). Specifically, each element \( e \) possesses 15 nodes with coordinates \( (X_{1}^{(e,m)}, X_{2}^{(e,m)}, X_{3}^{(e,m)}) \), \( m = 1, ..., 15 \). In terms of these nodal coordinates, with help of the reference tetrahedral element \( T =
{\(\xi, \eta, \rho\) : \(0 \leq \xi, \eta, \rho \leq 1\), \(\xi + \eta + \rho \leq 1\)}, the domain occupied by each element \(e\) is defined parametrically by \(E^{(e)} = \{(X_1, X_2, X_3) : X_i = M(\xi, \eta, \rho) = \sum_{m=1}^{10} N_Q^{(m)}(\xi, \eta, \rho) X_i^{(e,m)}, (\xi, \eta, \rho) \in T\}\). Figure D.1 illustrates a generic element \(E^{(e)}\) and its mapping from the reference element \(T\) indicating the location of all of its 15 nodes. In the above-introduced parametric mapping \(M\), \(N_Q^{(m)}(\xi, \eta, \rho)\) stand for the shape functions associated with the classical 10-node tetrahedral element.

For convenience, We recall them here:

\[
\begin{align*}
N_Q^{(1)}(\xi, \eta, \rho) &= \xi(2\xi - 1), & N_Q^{(2)}(\xi, \eta, \rho) &= \eta(2\eta - 1), \\
N_Q^{(3)}(\xi, \eta, \rho) &= (1 - \xi - \eta - \rho)(1 - 2\xi - 2\eta - 2\rho), & N_Q^{(4)}(\xi, \eta, \rho) &= \rho(2\rho - 1), \\
N_Q^{(5)}(\xi, \eta, \rho) &= 4\xi\eta, & N_Q^{(6)}(\xi, \eta, \rho) &= 4\eta(1 - \xi - \eta - \rho), & N_Q^{(7)}(\xi, \eta, \rho) &= 4\xi(1 - \xi - \eta - \rho), \\
N_Q^{(8)}(\xi, \eta, \rho) &= 4\xi\rho, & N_Q^{(9)}(\xi, \eta, \rho) &= 4\eta\rho, & N_Q^{(10)}(\xi, \eta, \rho) &= 4\rho(1 - \xi - \eta - \rho). \\
\end{align*}
\]

(D.31)

Figure D.1: Schematic of a “curved” tetrahedral element occupying a domain \(E^{(e)} \subseteq Y\) and its mapping \(M\) from the reference element \(T\). Blue circles indicate nodes 1 through 10. They are located at the four vertices and the six mid-edge points of \(T\). Nodes 11 through 14, which are located at the four mid-face points of \(T\), are shown as red circles. Node 15, located at the barycenter of \(T\), is indicated by a black square.

Having defined the partition of the unit cell \(Y\), we are now in a position to define \(U_h\), \(F_h\), and \(P_h\), the finite dimensional subspaces of \(U\), \(F\), and \(P\). They read as follows:

\[
\begin{align*}
U^h &= \{u^h \in [C^0(Y)]^3 \cap U : u^h_i(X_1, X_2, X_3)\}_{E^{(e)}} = \sum_{m=1}^{15} N_{CR}^{(m)}(\xi, \eta, \rho) u_i^{(e,m)}, \forall e = 1, ..., N_e\}, \\
F^h &= \{\Phi^h \in C^0(Y) \cap F : \Phi^h(X_1, X_2, X_3)\}_{E^{(e)}} = \sum_{m=1}^{15} N_{CR}^{(m)}(\xi, \eta, \rho) \Phi^{(e,m)}, \forall e = 1, ..., N_e\}, \\
P^h &= \{p^h \in P : p^h(X_1, X_2, X_3)\}_{E^{(e)}} = \sum_{k=0}^{3} N_p^{(k)}(\xi, \eta, \rho) p^{(e,k)}, \forall e = 1, ..., N_e\}. \\
\end{align*}
\]

(D.32)
Here, $C^0(Y)$ stands for the set of continuous functions defined on $Y$, $u_i^{(e,m)}$ and $\Phi^{(e,m)}$ denote the component $i$ of the displacement and the electric potential at node $m$ of element $e$, while $p^{(e,k)}$ denote the value ($k = 0$) of the pressure field and the three components ($k = 1, 2, 3$) of its gradient at node 15 of the domain $E^{(e)}$. In the above expressions, $N_{CR}^{(m)}(\xi, \eta, \rho)$ stand for the shape functions

\begin{align*}
N_{CR}^{(1)}(\xi, \eta, \rho) &= 3\xi[\eta\rho + (\eta + \rho)(1 - \eta - \xi - \rho)] + \xi[2\xi - 1 - 4\eta\rho(1 - \eta - \xi - \rho)], \\
N_{CR}^{(2)}(\xi, \eta, \rho) &= 3\eta[\xi\rho + (\xi + \rho)(1 - \eta - \xi - \rho)] + \eta[2\eta - 1 - 4\xi\rho(1 - \eta - \xi - \rho)], \\
N_{CR}^{(3)}(\xi, \eta, \rho) &= 3(1 - \eta - \xi - \rho)(\eta\xi + \eta\rho + \xi\rho) + (1 - \eta - \xi - \rho)[1 - 2\eta - 2\xi - 2\rho - 4\eta\xi\rho], \\
N_{CR}^{(4)}(\xi, \eta, \rho) &= 3\rho[\eta\xi + (\eta + \xi)(1 - \eta - \xi - \rho)] + \rho[2\rho - 1 - 4\eta\xi(1 - \eta - \xi - \rho)], \\
N_{CR}^{(5)}(\xi, \eta, \rho) &= 4\eta\xi[8\rho(1 - \eta - \xi - \rho) - 3(1 - \eta - \xi)] + 1], \\
N_{CR}^{(6)}(\xi, \eta, \rho) &= 4\eta(1 - \eta - \xi - \rho)[8\xi\rho - 3(\xi + \rho) + 1], \\
N_{CR}^{(7)}(\xi, \eta, \rho) &= 4\xi(1 - \eta - \xi - \rho)[8\eta\rho - 3(\eta + \rho) + 1], \\
N_{CR}^{(8)}(\xi, \eta, \rho) &= 4\rho[8\eta(1 - \eta - \xi - \rho) - 3(1 - \xi - \rho) + 1], \\
N_{CR}^{(9)}(\xi, \eta, \rho) &= 4\eta[8\xi(1 - \eta - \xi - \rho) - 3(1 - \eta - \rho) + 1], \\
N_{CR}^{(10)}(\xi, \eta, \rho) &= 4\rho(1 - \eta - \xi - \rho)[8\eta\xi - 3(\eta + \xi) + 1], \\
N_{CR}^{(11)}(\xi, \eta, \rho) &= 27\eta\xi(1 - 4\rho)(1 - \eta - \xi - \rho), \\
N_{CR}^{(12)}(\xi, \eta, \rho) &= 27\eta\rho(4\eta + 4\xi + 4\rho - 3), \\
N_{CR}^{(13)}(\xi, \eta, \rho) &= 27\eta\rho(1 - 4\xi)(1 - \eta - \xi - \rho), \\
N_{CR}^{(14)}(\xi, \eta, \rho) &= 27\xi\rho(1 - 4\rho)(1 - \eta - \xi - \rho), \\
N_{CR}^{(15)}(\xi, \eta, \rho) &= 256\eta\xi\rho(1 - \eta - \xi - \rho), \quad \text{(D.33)}
\end{align*}

while $N_{p}^{(k)}(\xi, \eta, \rho)$ are given by

\begin{align*}
N_{p}^{(0)}(\xi, \eta, \rho) &= 1, \quad N_{p}^{(1)}(\xi, \eta, \rho) = \xi - \frac{1}{4}, \quad N_{p}^{(2)}(\xi, \eta, \rho) = \eta - \frac{1}{4}, \quad N_{p}^{(3)}(\xi, \eta, \rho) = \rho - \frac{1}{4}. \quad \text{(D.34)}
\end{align*}

Again, the parameters $\xi, \eta, \rho$ in expressions (D.32)–(D.34) are related to the physical coordinates $X_1$, $X_2$, $X_3$ through the mapping $M$. At this stage, it is fitting to mention that the finite dimensional subspaces (D.32) and (D.33) for the displacement field $\mathbf{u}$ and the pressure field $p$ are three-dimensional analogues (see, e.g., Chapter II in Girault and Raviart, 1986; Chapter 8 in Boffi et al., 2012) of finite dimensional subspaces originally introduced in the context of two-dimensional Stokes flow by Crouzeix and Raviart (1973), thus the $CR$ subscript in (D.33). They have the merit to have been proven to lead to stable and convergent formulations in the context of isotropic linear elasticity,
irrespective of the compressibility of the medium (Chapter II in Girault and Raviart, 1986; Chapter 8 in Boffi et al., 2012). Moreover, in the context of nonlinear pdes, our own numerical investigations and those of others for different types of nonlinear pdes (see, e.g., van de Vosse, 2003) have shown that the finite dimensional subspaces (D.32) lead to formulations with the expected convergence rates, namely, in the present context, \(|u^h - u|_{L^2} = O(h^3), ||\Phi^h - \Phi||_{L^2} = O(h^3), ||p^h - p||_{L^2} = O(h^2)| with \(h = \max \text{diam}\{\mathcal{E}^{(c)}\}\). To avoid loss of continuity, details on the convergence properties for these FE formulations are deferred to Appendix D.4.2.

Standard assembly procedures allows one to construct global shape functions \(N_{CR}^{h(n)}(X), n = 1, \ldots, N_n\), and \(N_{P}^{h(l)}(X), l = 0, \ldots, 4N_e - 1\), so that the displacement \(u^h\) and the electric potential \(\Phi^h\) can be written in global form

\[
\begin{align*}
  u^h_i(X) &= \sum_{n=1}^{N_n} u^{(n)}_i N_{CR}^{h(n)}(X), \\
  \Phi^h(X) &= \sum_{n=1}^{N_n} \Phi^{(n)} N_{CR}^{h(n)}(X), \quad \text{and} \\
  p^h(X) &= \sum_{l=0}^{4N_e-1} p^{(l)} N_{P}^{h(l)}(X),
\end{align*}
\]

(D.35)

where the global degrees of freedom \(u^{(n)}_i\) and \(\Phi^{(n)}\) correspond physically to the component \(i\) of the displacement \(u^h\) and the electric potential \(\Phi^h\) at node \(n\), while \(p^{(l)}\) correspond physically to the value \((l \equiv 0 \mod 4)\) of the pressure field \(p^h\) and the three components \((l \equiv 1, 2, 3 \mod 4)\) of its gradient at the barycenter of \(\mathcal{E}^{(c)}\), \(e = [l/4] + 1\), \(l \not\equiv 0 \mod 4\); here, \([x] = \max \{z \in \mathbb{Z} : z \leq x\}\) denotes the floor function. Similarly, the test functions \(v^h \in U^h \cap U^0\), \(\psi^h \in \mathcal{F}^h \cap \mathcal{F}^0\), and \(q^h \in \mathcal{P}^h\) can be written in global form as

\[
\begin{align*}
  v^h_i(X) &= \sum_{n=1}^{N_n} v^{(n)}_i N_{CR}^{h(n)}(X), \\
  \psi^h(X) &= \sum_{n=1}^{N_n} \psi^{(n)} N_{CR}^{h(n)}(X), \quad \text{and} \\
  q^h(X) &= \sum_{l=0}^{4N_e-1} q^{(l)} N_{P}^{h(l)}(X).
\end{align*}
\]

(D.36)

In the above expressions, \(N_n\) stands for the total number of nodes in the partition of the cubic unit cell \(Y\). Their location must be selected so that the intersections of any two opposite faces of the partition of \(Y\) are identical. This geometric constraint is needed to be able to consistently enforce the required periodicity conditions (implied by relations (8.106)_{1-2}) of the continuous trial fields \(u^h\) and \(\Phi^h\) and the \(Y\)-periodicity of the continuous test functions \(v^h\) and \(\psi^h\). In light of this constraint, it proves useful to define the subsets of interior-node numbers \(I\), vertex-node numbers \(V\), face-node
numbers \( F \), and edge-node numbers \( E \) that make up the entire set of nodes. They read as

\[
I = \left\{ n : X_i^{(n)} \in (0, 1) \; \forall i \in \{1,2,3\} \right\}, \quad V = \left\{ n : X_i^{(n)} \in \{0,1\} \; \forall i \in \{1,2,3\} \right\},
\]

\[
F = \left\{ (n,p) : X_j^{(n)} = X_j^{(p)} \in (0,1), \; X_k^{(n)} = X_k^{(p)} \in (0,1), \; X_i^{(n)} = 0, \; X_i^{(p)} = 1 \; \forall (i,j,k) : \epsilon_{ijk} = 1 \right\},
\]

\[
E = \left\{ (n,p,q,r) : X_i^{(n)} = X_i^{(p)} = X_i^{(q)} = X_i^{(r)} \in (0,1), \; X_j^{(n)} = X_j^{(p)} = X_j^{(q)} = X_j^{(r)} = 0,
X_j^{(p)} = X_j^{(q)} = X_j^{(r)} = 1 \; \forall (i,j,k) : \epsilon_{ijk} = 1 \right\},
\]

(D.37)

where \( \epsilon_{ijk} \) stands for the three-dimensional permutation symbol.

Direct use of the trial fields (D.35) and the test functions (D.36) in the Euler-Lagrange equations
(8.107) leads to the following system of nonlinear algebraic equations

\[
\int_Y \left\{ p^h \left( \det F(u^h) \right) F^{-1}_{ji}(u^h) \frac{\partial \vec{W}^*}{\partial p}(X, F(u^h), p^h, E(\Phi^h)) \right\} \frac{\partial N^{h(n)}_{CR}}{\partial X_j}(X) dX = 0 \quad n \in I
\]

\[
\int_Y \left\{ p^h \left( \det F(u^h) \right) F^{-1}_{ji}(u^h) \frac{\partial \vec{W}^*}{\partial p}(X, F(u^h), p^h, E(\Phi^h)) \right\} \times \left( \frac{\partial N^{h(n)}_{CR}}{\partial X_j}(X) + \frac{\partial N^{h(p)}_{CR}}{\partial X_j}(X) + \frac{\partial N^{h(q)}_{CR}}{\partial X_j}(X) + \frac{\partial N^{h(r)}_{CR}}{\partial X_j}(X) \right) dX = 0 \quad (n, p) \in F
\]

\[
\int_Y \left\{ \frac{\partial \vec{W}^*}{\partial E_i}(X, F(u^h), p^h, E(\Phi^h)) \right\} \frac{\partial N^{h(n)}_{CR}}{\partial X_i}(X) dX = 0 \quad n \in I
\]

\[
\int_Y \left\{ \frac{\partial \vec{W}^*}{\partial E_i}(X, F(u^h), p^h, E(\Phi^h)) \right\} \left( \frac{\partial N^{h(n)}_{CR}}{\partial X_i}(X) + \frac{\partial N^{h(p)}_{CR}}{\partial X_i}(X) \right) dX = 0 \quad (n, p) \in F
\]

\[
\int_Y \left\{ \frac{\partial \vec{W}^*}{\partial E_i}(X, F(u^h), p^h, E(\Phi^h)) \right\} \times \left( \frac{\partial N^{h(n)}_{CR}}{\partial X_i}(X) + \frac{\partial N^{h(p)}_{CR}}{\partial X_i}(X) + \frac{\partial N^{h(q)}_{CR}}{\partial X_i}(X) + \frac{\partial N^{h(r)}_{CR}}{\partial X_i}(X) \right) dX = 0 \quad (n, p, q, r) \in E
\]

\[
\int_Y \left\{ \det F(u^h) - 1 - \frac{\partial \vec{W}^*}{\partial p}(X, F(u^h), p^h, E(\Phi^h)) \right\} N^{h(l)}_p(X) dX = 0 \quad l = 0, ..., 4N_e - 1
\]

(D.38)
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for the global degrees of freedom \( u^{(n)}_i \), \( \Phi^{(n)} \), \( p^{(l)} \) for \( n \notin \mathcal{V} \), together with the periodicity constraints

\[
\begin{align*}
    u^h_i(1, X_2, X_3) - u^h_i(0, X_2, X_3) &= (F_{i1} - \delta_{i1}), \\
    u^h_i(X_1, 1, X_3) - u^h_i(X_1, 0, X_3) &= (F_{i2} - \delta_{i2}), \\
    u^h_i(X_1, X_2, 1) - u^h_i(X_1, X_2, 0) &= (F_{i3} - \delta_{i3}), \\
    \Phi^h(1, X_2, X_3) - \Phi^h(0, X_2, X_3) &= -E_1, \\
    \Phi^h(X_1, 1, X_3) - \Phi^h(X_1, 0, X_3) &= -E_2, \\
    \Phi^h(X_1, X_2, 1) - \Phi^h(X_1, X_2, 0) &= -E_3,
\end{align*}
\]

and known values at the vertices

\[
\begin{align*}
    u^{(n)}_i &= (F_{ij} - \delta_{ij})X_j^{(n)} \quad \text{and} \quad \Phi^{(n)} = -E_iX_i^{(n)} \quad \text{for} \ n \in \mathcal{V}.
\end{align*}
\]

Having computed the values of all the degrees of freedom \( u^{(n)}_i \), \( \Phi^{(n)} \), \( p^{(l)} \) from the set of algebraic equations (D.38)–(D.40) for a given discretization of \( Y \), given local free-energy function \( W(X, F, E) \), and given electromechanical loading \( \mathbf{F} \) and \( \mathbf{E} \), the pointwise displacement field \( u^h(X) \), electric potential \( \Phi^h(X) \), and pressure field \( p^h(X) \) can be readily determined from expressions (D.35). In turn, these fields can be readily utilized to compute the effective free-energy function (8.105) for the dielectric elastomer composite of interest. Here, it is important to recall that the elastic dielectric response of dielectric elastomer composites may exhibit electromechanical limit loads. In order to be able to compute solutions past beyond eventual electromechanical limit loads, we solve the system of nonlinear algebraic equations (D.38)–(D.40) by means of an arc-length method. In particular, while there are many possibilities available, we found the use of the standard constraint condition put forward by Crisfield (1981) to be adequate for our purposes here.

D.4.2 Convergence study of the hybrid FE formulation

Next, we present the convergence properties of the FE formulation of the hybrid variational problem (8.105) based on the Crouzeix-Raviart type tetrahedral finite elements described above. To this end, we confront the FE solution to one of the rare physically meaningful solutions that can be determined in closed form within the context of finite electroelastostatics: the radially symmetric deformation of a spherical shell that is subjected to hydrostatic loading and a voltage on its boundaries. In its undeformed configuration, the shell is taken to occupy the domain \( \mathcal{S} = \{ X : A \leq |X| \leq B \} \) where \( 0 < A < B \). For definiteness, the shell is also assumed to be made up of a homogeneous isotropic...
ideal elastic dielectric characterized by the free energy

\[ W(F, E) = \frac{\mu}{2} [F \cdot F - 3] - \mu(J - 1) + \frac{\lambda + \mu}{2} (J - 1)^2 - \frac{\varepsilon}{2} J F^{-T} E \cdot F^{-T} E, \quad (D.41) \]

where we recall the definition \( J = \det F \) and that \( \mu, \lambda, \varepsilon \) stand for the initial Lamé constants and permittivity of the material. In this context, by analogy with (8.105) and (8.103), we have the hybrid variational problem

\[
\min_{u \in U} \max_{\Phi \in \mathcal{F}} \max_{p \in \mathcal{P}} \int_S \left\{ p[\det F(u) - 1] - \hat{W}^*(F(u), p, E(\Phi)) \right\} \, dX, \quad (D.42)
\]

where

\[
\hat{W}^*(F, p, E) = p - \frac{\mu}{2} [F \cdot F - 3] + \frac{\mu - \lambda}{2} + \varepsilon F^{-T} E \cdot F^{-T} E + \frac{(\varepsilon F^{-T} E \cdot F^{-T} E + 2p - 2\lambda)^2}{8(\lambda + \mu)}, \quad (D.43)
\]

and \( U, \mathcal{F}, \) and \( \mathcal{P} \) stand for sufficiently large sets of admissible displacements \( u \), admissible electric potentials \( \Phi \), and admissible pressure fields \( p \) that are consistent with the choice of boundary conditions

\[
(p \det F) F^{-T} X - \frac{\partial W^*}{\partial F} X = 0, \quad \Phi = \Phi_A \quad \text{for} \quad |X| = A \quad (D.44)
\]

and

\[
u = (\lambda - 1) X, \quad \Phi = \Phi_B \quad \text{for} \quad |X| = B. \quad (D.45)
\]

The above boundary conditions correspond physically to taking the inner boundary of the shell to be traction free, applying a radial displacement on the outer boundary of the shell, and prescribing their voltage to be \( \Phi_A \) and \( \Phi_B \), respectively. The Euler-Lagrange equations associated with (D.42) are given by

\[
\text{Div} \left[ (p \det F) F^{-T} - \frac{\partial W^*}{\partial F} \right] = 0, \quad \text{Div} \frac{\partial W^*}{\partial E} = 0, \quad \det F - 1 - \frac{\partial W^*}{\partial p} = 0. \quad (D.46)
\]

**D.4.2.1 Closed-form radially symmetric solution of (D.42)**

In order to construct the closed-form radially symmetric solution for the Euler-Lagrange equations (D.46), we write

\[
u = \left( \frac{r(R)}{R} - 1 \right) X, \quad \Phi = \Phi(R), \quad \text{and} \quad p = p(R), \quad (D.47)
\]
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where \( R \equiv |\mathbf{X}| \) and the functions \( r(R), \Phi(R), \) and \( p(R) \) are to be determined. For this class of fields, the deformed configuration of the shell is then defined as \( \mathcal{S}_d = \{ \mathbf{x} : a \leq |\mathbf{x}| \leq b \} \) where \( a = r(A) \) and \( b = r(B) = \lambda B \) from (D.45).1.

Because the focus of Chapters 7 and 8 is on incompressible dielectric elastomer composites, we consider in the rest of this appendix that the shell is made up of an incompressible material. In the limit \( \lambda/\mu \to \infty \), (D.43) reduces to

\[
\hat{W}^*(\mathbf{F}, p, \mathbf{E}) = -\frac{\mu}{2} |\mathbf{F} \cdot \mathbf{F} - 3| + \frac{\varepsilon}{2} \mathbf{F}^{-T} \mathbf{E} \cdot \mathbf{F}^{-T} \mathbf{E}.
\] (D.48)

We remark already that in this case, the Euler-Lagrange equation (D.46) is equivalent to the local incompressibility constraint \( \det \mathbf{F} = 1 \). This constraint allows to determine analytically the function \( r(R) \) as

\[
r(R) = \left( 1 + \frac{b^3 - B^3}{R^3} \right)^{1/3} R.
\] (D.49)

It follows that

\[
\mathbf{u} = \left( 1 + \frac{b^3 - B^3}{R^3} \right)^{1/3} \mathbf{X} - \mathbf{X} \quad \text{and} \quad a = \left( 1 + \frac{b^3 - B^3}{A^3} \right) A.
\] (D.50)

After recognizing that \( \mathbf{F} \) and \( \mathbf{E} \) can be expressed in the spectral forms

\[
\mathbf{F} = \frac{1}{r(R)^2} \mathbf{X} \otimes \mathbf{X} + \frac{r(R)}{R} \left( \mathbf{I} - \frac{1}{R^2} \mathbf{X} \otimes \mathbf{X} \right) \quad \text{and} \quad \mathbf{E} = -\Phi'(R) \frac{\mathbf{X}}{R},
\] (D.51)

with \( \Phi'(R) \equiv d\Phi/dR \), the Euler-Lagrange equation (D.46) reduces here to the second-order ordinary differential equation (ode)

\[
R(R^3 + b^3 - B^3)\Phi''(R) + 2(R^3 + B^3 - b^3)\Phi'(R) = 0,
\] (D.52)

with \( \Phi''(R) \equiv d^2\Phi/dR^2 \). With help of the boundary conditions (D.44) and (D.45), it is a simple matter to deduce that

\[
\Phi = -\frac{ab(\Phi_B - \Phi_A)}{(b - a)(b^3 - B^3 + R^3)^{1/3}} + \frac{b\Phi_B - a\Phi_A}{b - a}
\] (D.53)

where \( a \) is given by (D.50) and \( b = \lambda B \). Similarly, upon recognizing that the Cauchy stress

\[
\mathbf{T} = p\mathbf{I} - \frac{1}{\det \mathbf{F}} \frac{\partial \hat{W}^*}{\partial \mathbf{F}} \mathbf{F}^T
= p\mathbf{I} + \mu \mathbf{F} \mathbf{F}^T + \varepsilon \mathbf{F}^{-T} \mathbf{E} \otimes \mathbf{F}^{-T} \mathbf{E}
\] (D.54)
admits the spectral form

$$T = \frac{t_1(R)}{R^2} \mathbf{X} \otimes \mathbf{X} + t_2(R) \left( \mathbf{I} - \frac{1}{R^2} \mathbf{X} \otimes \mathbf{X} \right), \quad (D.55)$$

(D.46) can be shown to reduce to the first-order ode

$$t_1'(R) = \frac{2R^2}{r(R)^3} [t_2(R) - t_1(R)] \quad (D.56)$$

with \( t_1'(R) = dt_1/dR \) and \( r(R) \) given by (D.49). It follows from (D.51), (D.54), and (D.55) that

$$t_2(R) - t_1(R) = \left( b^3 - B^3 \right) \left( b^3 - B^3 + 2R^3 \right) \mu \frac{a^2 b^2 \varepsilon (\Phi_B - \Phi_A)^2}{(b - a)^2(b^3 - B^3 + R^3)^{4/3}}. \quad (D.57)$$

With help the boundary condition (D.44), using the above expression into (D.56) renders analytical expressions for \( t_1(R) \) and subsequently for \( t_2(R) \). Because of their bulkiness, we do not report them here. However, upon noting from (D.54) that

$$p = \frac{t_1(R) + 2t_2(R)}{3} - \frac{\mu}{3} \mathbf{F} \cdot \mathbf{F} - \frac{\varepsilon}{3} \mathbf{F}^{-T} \mathbf{E} \cdot \mathbf{F}^{-T} \mathbf{E}, \quad (D.58)$$

and making use of the above-described expressions for \( t_1(R) \) and \( t_2(R) \), the pressure-like field \( p \) reads finally as

$$p = \int_A \frac{2 \left( b^3 - B^3 \right) \left( b^3 - B^3 + 2z^3 \right) \mu}{(b^3 - B^3 + z^3)^{7/3}} \left[ \frac{a^2 b^2 \varepsilon (\Phi_B - \Phi_A)^2}{(b - a) \left( b^3 - B^3 + z^3 \right)^{7/3}} \right] \, dz - \frac{R^4 \mu}{(b^3 - B^3 + R^3)^{4/3}} - \frac{a^2 b^2 \varepsilon (\Phi_B - \Phi_A)^2}{(b - a)^2 \left( b^3 - B^3 + R^3 \right)^{4/3}}, \quad (D.59)$$

with, again, \( a \) given by (D.50) and \( b = \bar{\lambda}B \).

### D.4.2.2 Convergence properties of the hybrid FE discretization

Equipped with the analytical solutions (D.50), (D.53), and (D.59) of the variational problem (D.42) when the shell is made out of an incompressible elastic dielectric, we finally report the convergence properties of the hybrid FE discretization of (D.42) based on the Crouzeix-Raviart type tetrahedral finite-element described in Appendix D.4.1. The construction of the trial fields \( \mathbf{u}^h, \Phi^h, \) and \( p^h \) are similar to the one outlined in Appendix D.4.1 and hence is not reported here.

The convergence study was conducted with the following choice of values for the parameters entering (D.42): \( \mu = 1 \text{ MPa}, \lambda/\mu = 10^8, \varepsilon = 30.0 \times 10^{-12} \text{ F/m}, A = 0.1 \text{ mm}, B = 0.5 \text{ mm}, \bar{\lambda} = 1.01, \Phi_A = 0 \text{ V}, \Phi_B = 15 \text{ V}. \) In this context, the evolution with the maximum element diameter \( h \) of the normalized \( L^2 \)-norms of the errors in (a) the displacement field \( \mathbf{u}^h - \mathbf{u} \), (b) the
electric potential $\Phi_h - \Phi$, and (c) the pressure-like field $p_h - p$ are presented of Fig. D.2. Figure D.2 shows that the numerical solutions converge to the analytical results (D.50), (D.53), and (D.59) as $\|u_h - u\|_{L^2} \approx O(h^{2.5}), \|\Phi_h - \Phi\|_{L^2} \approx O(h^{2.8}), \|p_h - p\|_{L^2} \approx O(h^{1.6})$ with the mesh refinement. These results are in fairly good agreement with the expected convergence rates $\|u_h - u\|_{L^2} = O(h^3), \|\Phi_h - \Phi\|_{L^2} = O(h^3), \|p_h - p\|_{L^2} = O(h^2)$.

Figure D.2: Evolution of the normalized $L^2$-norms of the errors in (a) the displacement field $u_h - u$, (b) the electric potential $\Phi_h - \Phi$, and (c) the pressure-like field between the analytical solutions (D.50), (D.53), and (D.59) of the variational problem (D.42), and the FE solutions obtained via the hybrid FE discretization of based on the Crouzeix-Raviart type tetrahedral finite-element described in Appendix D.4.1.
Governing equations for elastic dielectric composites in the limit of small deformations and moderate electric fields in the presence of space charges

We derive in this appendix the governing equations for elastic dielectric composites in the limit of small deformations and moderate electric fields in the presence of space charges. In addition to supplementing Chapter 9 where elastic dielectric composites comprising space charges are addressed, this appendix serves as a complement to Chapter 2 where the governing equations for elastic dielectric composites in the absence of space charges were initially presented.

Consider a deformable composite material with periodic microstructure of period $\delta$ that occupies a bounded domain $\Omega \subset \mathbb{R}^N$ ($N = 1, 2, 3$), with smooth boundary $\partial \Omega$ and closure $\overline{\Omega} = \Omega \cup \partial \Omega$, in its undeformed configuration. Material points are identified by their initial position vector $X$ in $\Omega$ relative to some fixed point. Upon the application of mechanical and electrical stimuli, the position vector $X$ of a material point moves to a new position specified by $x = X + u^\delta(X)$, where the displacement field $u^\delta(X)$ is loosely taken to possess sufficient regularity to warrant the mathematical well-posedness of the equations that follow. The associated deformation gradient is denoted by $F^\delta(X) = I + \text{Grad } u^\delta(X)$.

In the absence of magnetic fields, free currents, and body forces, and with no time dependence (see, e.g., Chapter 15 in Kovetz, 2000; see also Dorfmann and Ogden, 2005a; Lopez-Pamies, 2014),
Maxwell’s and momentum balance equations require that

\[ \text{Div} \mathbf{D}^\delta(\mathbf{x}) = q^\delta(\mathbf{x}), \quad \text{Curl} \mathbf{E}^\delta(\mathbf{x}) = 0, \quad \mathbf{x} \in \mathbb{R}^N \quad (E.1) \]

and

\[ \text{Div} \mathbf{S}^\delta(\mathbf{x}) = 0, \quad \mathbf{S}^\delta \mathbf{F}^T = \mathbf{F}^\delta \mathbf{S}^T, \quad \mathbf{x} \in \Omega, \quad (E.2) \]

where \( \mathbf{D}^\delta(\mathbf{x}), \mathbf{E}^\delta(\mathbf{x}), \mathbf{S}^\delta(\mathbf{x}) \) stand for the Lagrangian electric displacement field, the Lagrangian electric field, and the “total” first Piola-Kirchhoff stress tensor, while \( q^\delta(\mathbf{x}) \) stands for the density of space charges per unit undeformed volume. For the specific case when the composite material is a (hyper-)elastic dielectric with even electromechanical coupling, we further have the constitutive connections

\[ \mathbf{D}^\delta(\mathbf{x}) = -\frac{\partial W^\delta}{\partial \mathbf{E}^\delta}(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta) \quad \text{and} \quad \mathbf{S}^\delta(\mathbf{x}) = \frac{\partial W^\delta}{\partial \mathbf{F}^\delta}(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta), \quad (E.3) \]

where the “total” free energy \( W^\delta(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta) \) is an objective function of the deformation gradient tensor \( \mathbf{F}^\delta \) and an even and objective function of the electric field \( \mathbf{E}^\delta \), namely, \( W^\delta(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta) = W^\delta(\mathbf{x}, \mathbf{Q} \mathbf{F}^\delta, \mathbf{E}^\delta) = W^\delta(\mathbf{x}, \mathbf{F}^\delta, -\mathbf{E}^\delta) \) for all \( \mathbf{Q} \in \text{Orth}^+ \) and arbitrary \( \mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta \).

Upon recognizing that the assumed objectivity of \( W^\delta(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta) \) implies the automatic satisfaction of the balance of angular momentum \((E.2)_2\) and that Faraday’s law \((E.1)_2\) is automatically satisfied by the introduction of an electric potential \( \varphi^\delta(\mathbf{x}) \), taken to possess appropriate regularity, such that

\[ \mathbf{E}^\delta(\mathbf{x}) = -\text{Grad} \varphi^\delta(\mathbf{x}), \quad (E.4) \]

the equations governing the elastic dielectric response of the composite material reduce to the pdes

\[ \text{Div} \left[ -\frac{\partial W^\delta}{\partial \mathbf{E}^\delta}(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta) \right] = q^\delta(\mathbf{x}), \quad \mathbf{x} \in \mathbb{R}^N \quad \text{and} \quad \text{Div} \left[ \frac{\partial W^\delta}{\partial \mathbf{F}^\delta}(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta) \right] = 0, \quad \mathbf{x} \in \Omega. \quad (E.5) \]

The classical limit of small deformations and moderate electric fields  Now, let us define \( \zeta \) as a vanishingly small parameter and take the deformation measure \( \mathbf{H}^\delta(\mathbf{x}) = \mathbf{F}^\delta(\mathbf{x}) - \mathbf{I} = \text{Grad} \mathbf{u}^\delta(\mathbf{x}) \) to be \( O(\zeta) \) and the electric field \( \mathbf{E}^\delta(\mathbf{x}) = -\text{Grad} \varphi^\delta(\mathbf{x}) \) to be \( O(\zeta^{1/2}) \). Then, assuming that the composite material is stress-free in the undeformed configuration \( \Omega \), the asymptotic result

\[ W^\delta(\mathbf{x}, \mathbf{F}^\delta, \mathbf{E}^\delta) = -\frac{1}{2} \mathbf{E}^\delta_i \varphi^\delta_\delta(\mathbf{x}) \mathbf{E}^\delta_j + \frac{1}{2} H^\delta_{ij} L^\delta_{ijkl}(\mathbf{x}) H^\delta_{kl} + H^\delta_{ij} M^\delta_{ijkl}(\mathbf{x}) E^\delta_k E^\delta_l - E^\delta_i E^\delta_j T^\delta_{ijkl}(\mathbf{x}) E^\delta_k E^\delta_l + O(\zeta^3) \quad (E.6) \]
follows from a simple formal calculation (and the physically inconsequential choice that $W^\delta(X, I, 0) = 0$). Here, $\varepsilon^\delta_{ij}(X) = -\partial^2 W^\delta(X, I, 0)/\partial E^\delta_i \partial E^\delta_j$ is the permittivity tensor, $L^\delta_{ijkl}(X) = \partial^2 W^\delta(X, I, 0)/\partial F^\delta_{ij} \partial F^\delta_{kl}$ is the elasticity tensor, $M^\delta_{ijkl}(X) = 1/2 \partial^3 W^\delta(X, I, 0)/\partial F^\delta_{ij} \partial E^\delta_k \partial E^\delta_l$ is the electrostriction tensor, and $T^\delta_{ijkl}(X) = -1/24 \partial^4 W^\delta(X, I, 0)/\partial E^\delta_i \partial E^\delta_j \partial E^\delta_k \partial E^\delta_l$ is the permittivity tensor of second order. In turn, to leading order, the constitutive relations (E.3) reduce to (see also Chapter 2.25 in Stratton, 1941; Section 15 in Toupin, 1956; Chapters 2 and 3 in Tian, 2007; Schröder and Keip, 2012; Chapter 3):

$$D^\delta_i(X) = \varepsilon^\delta_{ij}(X) E^\delta_j + O(\zeta^{3/2}) \quad \text{and} \quad S^\delta_{ij}(X) = L^\delta_{ijkl}(X) H^\delta_{kl} + M^\delta_{ijkl}(X) E^\delta_k E^\delta_l + O(\zeta^2).$$  \hspace{1cm} (E.7)

By taking the space-charge density $q^\delta(X)$ to be $O(\zeta^{1/2})$, and by restricting attention to Dirichlet boundary conditions, the one-way coupled boundary-value problems (9.8)–(9.9) in the main body of the text follow readily upon direct use of the asymptotic expressions (E.7) in the governing pdes (E.5).
Comparison of the general homogenization result (10.30) with an existing result for circular rigid particles

In this appendix, we confront the result of Galipeau and Ponte Castañeda (2013) — the only existing (as far as the authors know) analytical homogenization estimate in the literature for the macroscopic magnetoelastic response of isotropic magnetorheological elastomers at finite deformations and finite magnetic fields — to the general homogenization result put forth in Chapter 10. As outlined in the introductory paragraphs of Chapter 10, the result of these authors corresponds to an approximation in $N = 2$ space dimensions for the effective Helmholtz free-energy function of an isotropic suspension of magnetizable rigid circular particles firmly embedded in an isotropic incompressible rubber matrix. For simplicity, we restrict the comparison to the fundamental limiting case of particles composed of a linear magnetic material, for which their result can be expressed explicitly.

In the notation/terminology employed in Chapter 10, taking the rubber matrix to be characterized by the free-energy function (10.1) and the isotropically distributed circular particles to be characterized by the free-energy function (10.2) with the material parameter $G_p = +\infty$ and the material function $S(I_5^H) = \mu_p I_5^H / 2$, it is not difficult to deduce that equations (19), (27), (30) in (Galipeau and Ponte Castañeda, 2013) can be combined and manipulated algebraically to yield the
F. Comparison of the general homogenization result (10.30) with an existing result

The effective Helmholtz free-energy function

\[ W^*(F, B) = \begin{cases} 
(1 - c)\Psi \left( \hat{T}_1 \right) - \frac{2c^2}{\mu_0} \left( \frac{\mu_p - \mu_0}{\mu_0(\alpha + \beta \hat{T}_1)} \right)^2 T_4^B + \frac{(\mu_p + \mu_0)^2}{2\mu_0} \left( \frac{2 + \hat{T}_1}{\alpha + \beta \hat{T}_1} \right) T_5^B & \text{if } \mathcal{J} = 1 \\
+\infty & \text{otherwise}
\end{cases} \quad (F.1) \]

with

\[ \alpha = 2[(1 + c)\mu_p + (1 - c)\mu_0]^2 + 2c^2(\mu_p - \mu_0)^2, \quad \beta = (\mu_p + \mu_0) [(1 + 2c)\mu_p + (1 - 2c)\mu_0], \]

\[ \hat{T}_1 = \frac{1 + 2c - 4c\hat{\lambda}}{(1 - c)^2\hat{\lambda}^2} + \frac{2c^2 - 4c\hat{\lambda} + (1 + 2c)^2}{(1 - c)^2}, \quad \hat{\lambda} = \sqrt{\frac{T_1 + \sqrt{T_1^2 - 4}}{2}}, \quad (F.2) \]

explicitly in terms of the standard invariants \( T_1, T_4^B, T_5^B \) defined by relations (10.15)_1 and (10.32)_1,2 in the main body of the text. By contrast, the effective Helmholtz free-energy function that ensues from the corresponding specialization of the general result (10.30) put forth in Chapter 10 — making use of the analytical coefficients (10.33) — is given by

\[ W^*(F, B) = \begin{cases} 
(1 - c)\Psi \left( \hat{T}_1^{\text{Cir.}} \right) + \frac{1}{2\tilde{n}} \left[ \tilde{\eta} T_4^B + T_5^B \left( 1 + \tilde{\eta}^2 + \tilde{\eta} T_1 \right) \right] & \text{if } \mathcal{J} = 1 \\
+\infty & \text{otherwise}
\end{cases} \quad (F.3) \]

where

\[ \tilde{\eta} = \frac{\tilde{\nu} - \tilde{n}}{\tilde{n}}, \quad \tilde{\nu} = \mu_0 + \frac{2c\mu_0(\mu_p - \mu_0)}{(1 + c)\mu_0 + (1 - c)\mu_p}, \]

\[ \tilde{n} = \mu_0 + \frac{c(8 + 4c + 3c^2 + c^3)(\mu_p - \mu_0)^2}{4[(1 - c)\mu_p + (1 + c)\mu_0]^2} + \frac{c(1 - c)(8 + 4c + c^2)(\mu_p - \mu_0)\mu_0\mu_p}{4[(1 - c)\mu_p + (1 + c)\mu_0]^2}, \]

\[ T_1^{\text{Cir.}} = \frac{T_1 - 2}{(1 - c)^3} + 2. \quad (F.4) \]

Interestingly, the estimate (F.1) of Galipeau and Ponte Castañeda (2013) is in qualitative agreement with the result (F.3) in that is linear in the invariants \( T_4^B \) and \( T_5^B \). Its dependence on the invariant \( \hat{T}_1 \) is different, however. Numerical comparisons indicate that the estimate (F.1) is in fair quantitative agreement with the result (F.3) for small volume fractions of particles and small deformations and magnetic fields. Quantitative differences do appear otherwise.