Nonlinear electroelastic deformations of dielectric elastomer composites: I — Ideal elastic dielectrics

Victor Lefèvre, Oscar Lopez-Pamies

Department of Civil and Environmental Engineering, University of Illinois, Urbana–Champaign, IL 61801, USA

Abstract

This paper 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. As elaborated in a companion paper, the proposed approximate solution proves particularly useful as a fundamental building block to generate approximate solutions for the macroscopic elastic dielectric response of dielectric elastomer composites made up of non-Gaussian dielectric elastomers filled with nonlinear elastic dielectric particles.

Key words: Iterated homogenization; Viscosity solution; Microstructures; Electroactive materials; Electrostriction

1. Introduction and main result

Since the turn of the millennium, dielectric elastomer composites — specifically, dielectric elastomers filled with (semi-)conducting or high-permittivity particles — have received increasing attention by the materials research community because of their potential to outperform unfilled dielectric elastomers for employment in emerging technologies (see, e.g., Zhang et al., 2002; Huang and Zhang, 2004; Huang et al., 2005; Carpi and Rossi, 2005; McCarthy et al., 2009; Meddeb and Ouanies, 2012; Liu et al., 2013). At present, however, the microscopic mechanisms responsible for the superior electromechanical properties of this type of electroactive composite materials remain unresolved. In the literature, there are two mechanisms that have been identified as possibly dominant: *i*) the nonlinear elastic dielectric nature of elastomers which heightens the role of the fluctuations of the electric field in the presence of filler particles (Li, 2003; Tian et al., 2012) and *ii*) the presence of high-dielectric interphases and/or interphasial free charges surrounding the filler particles (Lewis, 2004; Lopez-Pamies et al., 2014).

The objective of this two-part paper is to investigate the first of the two mechanisms stated above in the context of nonlinear electroelastic deformations. That is, we view dielectric elastomer composites as two-phase particulate composites — comprising a continuous dielectric elastomer matrix filled by a statistically uniform distribution of firmly bonded inclusions — and study their homogenized (macroscopic or overall) elastic dielectric response when subjected to finite deformations and finite electric fields. In light of the fact that the majority of existing experimental evidence pertains to dielectric elastomers filled with particles

Email addresses: vllefevre@illinois.edu (Victor Lefèvre), pamies@illinois.edu (Oscar Lopez-Pamies)

of roughly spherical shape that are distributed randomly without a biased direction, we shall focus our attention throughout this work on dielectric elastomer composites with isotropic microstructures.

To put the problem at hand in perspective, it is fitting to mention that within the classical asymptotic context of small deformations and moderate electric fields (see, e.g., Section 2.25 in Stratton, 1941) it is only about a decade ago that heuristic estimates for the macroscopic elastic dielectric response of dielectric elastomer composites were first reported (Li and Rao, 2004; Li et al., 2004); see also Siboni and Ponte Castañeda (2013) for the specific case when the particles are mechanically rigid. It was in a later contribution that Tian (2007) and Tian et al. (2012) established rigorously via a two-scale convergence analysis the homogenization limit of the equations of elastic dielectrics with *even* electroelastic coupling, the intrinsic coupling of dielectric elastomers. Remarkably, in spite of the coupling and nonlinearity of the underlying equations, their result indicates that the problem reduces to a system of two uncoupled linear partial differential equations (pdes). Lefèvre and Lopez-Pamies (2014) solved these pdes analytically and worked out closed-form formulas for the macroscopic elastic dielectric properties of dielectric elastomers filled with an isotropic distribution of polydisperse spherical particles of infinitely many sizes, the so-called differential coated sphere assemblage. Corresponding results for dielectric elastomers filled with an isotropic distribution of monodisperse spherical particles were later provided by Spinelli et al. (2015), who solved the pertinent pdes numerically by means of finite elements. Spinelli et al. (2015) also provided closed-form formulas for the macroscopic elastic dielectric properties of dielectric elastomer composites with a special class of isotropic iterative particulate microstructure due to Lopez-Pamies (2014).

In contrast to the above-outlined body of asymptotic work, results for finite deformations and finite electric fields have not been developed to nearly the same advanced state, presumably because of the inherent technical difficulties. Essentially, there are the analytical results of Lopez-Pamies (2014) for the overall electrostriction of an ideal elastic dielectric composite with the aforementioned isotropic iterative particulate microstructure. Albeit not for isotropic microstructures, there are also the recent finite-element results of Miehe et al. (2015) for the overall electrostriction of a *compressible* ideal elastic dielectric composite wherein the fillers are spherical particles distributed in a periodic cubic array.

The focus of this Part I of the work is on dielectric elastomer composites wherein the underlying matrix and particles are both ideal elastic dielectrics. The formulation of this fundamental problem is presented in Section 2. In the spirit of the strategy followed by Lefèvre and Lopez-Pamies (2014) and Spinelli et al. (2015), we then construct solutions for the macroscopic elastic dielectric response of ideal elastic dielectric composites with three different isotropic microstructures: *i*) the isotropic iterative microstructure of Lopez-Pamies (2014) 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. Section 3 presents the solution for the iterative microstructure, which happens to correspond to the viscosity solution of a Hamilton-Jacobi (HJ) pde in five “space” variables. Its computation is carried out by means of a novel weighted-essentially-non-oscillatory (WENO) finite-difference scheme (Lefèvre et al., 2016). Section 4 presents the solutions for the microstructures wherein the fillers are spherical particles of poly- and mono-disperse sizes. These are computed by means of hybrid finite elements. The results of Sections 3 and 4 prompt an approximate closed-form solution of remarkable simplicity for the macroscopic elastic dielectric response of ideal elastic dielectric composites with any type of non-percolative isotropic particulate microstructure. We describe in Sections 5 and 6 the features and physical implications of this approximation, but also record it here for convenience:

The macroscopic elastic dielectric response of an ideal elastic dielectric with (incompressible) free-energy function $W_m = \mu/2 [\mathbf{F} \cdot \mathbf{F} - 3] - \varepsilon/2 \mathbf{F}^{-T} \mathbf{E} \cdot \mathbf{F}^{-T} \mathbf{E}$, filled with any type of isotropic distribution of ideal elastic dielectric particles with (incompressible) free-energy function $W_p = \mu_p/2 [\mathbf{F} \cdot \mathbf{F} - 3] - \varepsilon_p/2 \mathbf{F}^{-T} \mathbf{E} \cdot \mathbf{F}^{-T} \mathbf{E}$, is characterized by the (incompressible) effective free-energy function

$$\overline{W} = \frac{\tilde{\mu}}{2} [\overline{\mathbf{F}} \cdot \overline{\mathbf{F}} - 3] + \frac{\tilde{m}_K - \tilde{\varepsilon}}{2} \overline{\mathbf{E}} \cdot \overline{\mathbf{E}} - \frac{\tilde{m}_K}{2} \overline{\mathbf{F}}^{-T} \overline{\mathbf{E}} \cdot \overline{\mathbf{F}}^{-T} \overline{\mathbf{E}}.$$

Here, $\overline{\mathbf{F}}$ and $\overline{\mathbf{E}}$ denote the macroscopic deformation gradient and macroscopic Lagrangian electric field, while the coefficients $\tilde{\mu}$, $\tilde{\varepsilon}$, $\tilde{m}_K$ stand for the effective shear modulus, effective permittivity, and effective electrostrictive constant that characterize the response of the composite material in the asymptotic limit of small deformations and moderate electric fields (see Section 2.1 below).

In a companion paper (Lefèvre and Lopez-Pamies, 2016), henceforth referred to as Part II, the above basic result for $\overline{W}$ will be utilized as a fundamental building block to generate approximate solutions for the macroscopic elastic dielectric response of dielectric elastomer composites comprised of non-Gaussian elastomers filled with nonlinear elastic dielectric particles that may exhibit polarization saturation.

2. The problem

Microscopic description of dielectric elastomer composites. A dielectric elastomer composite is taken here to consist of a distribution of filler particles firmly bonded to an otherwise homogeneous dielectric elastomer matrix. The domain occupied by the entire composite in its ground state is denoted by Ω and its boundary by $\partial\Omega$. Similarly, Ω_m and Ω_p denote the domains occupied collectively by the matrix and the particles so that $\Omega = \Omega_m \cup \Omega_p$. The initial volume fraction (or concentration) of particles is denoted by $c \doteq |\Omega_p|/|\Omega|$. We assume that the distribution of the particles is statistically uniform (i.e., translation invariant) and that their sizes are much smaller than the size of Ω . For convenience, we choose units of length so that Ω has unit volume.

Each material point in the initial configuration Ω is identified by its initial position vector $\mathbf{X}$, while its position in the deformed configuration is given by $\mathbf{x} = \boldsymbol{\chi}(\mathbf{X})$. We assume that the mapping $\boldsymbol{\chi}$ is bijective and twice continuously differentiable, except possibly on the boundaries between the particles and the matrix where it is only required to be continuous. The corresponding deformation gradient is denoted by $\mathbf{F} = \text{Grad } \boldsymbol{\chi}$ and its determinant by $J = \det \mathbf{F}$.

Both the matrix and the particles are assumed to be elastic dielectrics. We find it convenient to characterize their electromechanical behaviors by “total” free-energy functions (see, e.g., Dorfmann and Ogden, 2005) of the deformation gradient $\mathbf{F}$ and the Lagrangian electric field $\mathbf{E}$: $W_m = W_m(\mathbf{F}, \mathbf{E})$ and $W_p = W_p(\mathbf{F}, \mathbf{E})$. It follows that at each material point $\mathbf{X} \in \Omega$ the first Piola-Kirchhoff stress tensor $\mathbf{S}$ and Lagrangian electric displacement field $\mathbf{D}$ are formally given in terms of $\mathbf{F}$ and $\mathbf{E}$ by

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X}, \mathbf{F}, \mathbf{E}) \quad \text{and} \quad \mathbf{D} = -\frac{\partial W}{\partial \mathbf{E}}(\mathbf{X}, \mathbf{F}, \mathbf{E}), \quad (1)$$

where

$$W(\mathbf{X}, \mathbf{F}, \mathbf{E}) = [1 - \theta(\mathbf{X})]W_m(\mathbf{F}, \mathbf{E}) + \theta(\mathbf{X})W_p(\mathbf{F}, \mathbf{E}) \quad \text{with} \quad \theta(\mathbf{X}) = \begin{cases} 1 & \text{if } \mathbf{X} \in \Omega_p \\ 0 & \text{otherwise} \end{cases}. \quad (2)$$

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 $\overline{\mathbf{S}} \doteq \int_{\Omega} \mathbf{S}(\mathbf{X}) d\mathbf{X}$ and Lagrangian electric displacement field $\overline{\mathbf{D}} \doteq \int_{\Omega} \mathbf{D}(\mathbf{X}) d\mathbf{X}$ 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 Ω 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:

$$\mathbf{x} = \overline{\mathbf{F}}\mathbf{X} \quad \text{and} \quad \Phi = -\overline{\mathbf{E}} \cdot \mathbf{X} \quad \text{on} \quad \partial\Omega, \quad (3)$$

where the second-order tensor $\overline{\mathbf{F}}$ and vector $\overline{\mathbf{E}}$ stand for prescribed boundary data. It follows from the divergence theorem that $\int_{\Omega} \mathbf{F}(\mathbf{X}) d\mathbf{X} = \overline{\mathbf{F}}$ and $\int_{\Omega} \mathbf{E}(\mathbf{X}) d\mathbf{X} = \overline{\mathbf{E}}$, and hence that the sought macroscopic constitutive relation can be simply written as (Lopez-Pamies, 2014)

$$\overline{\mathbf{S}} = \frac{\partial \overline{W}}{\partial \overline{\mathbf{F}}}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, c) \quad \text{and} \quad \overline{\mathbf{D}} = -\frac{\partial \overline{W}}{\partial \overline{\mathbf{E}}}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, c), \quad (4)$$

where $\overline{W}$ corresponds physically to the total electroelastic free energy (per unit undeformed volume) of the composite as defined by

$$\overline{W}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, c) = \min_{\mathbf{F} \in \mathcal{K}} \max_{\mathbf{E} \in \mathcal{E}} \int_{\Omega} W(\mathbf{X}, \mathbf{F}, \mathbf{E}) d\mathbf{X}. \quad (5)$$

In these last expressions, the explicit dependence of the effective free-energy function $\overline{W}$ on the volume fraction of particles c has been introduced for later convenience and $\mathcal{K}$, $\mathcal{E}$ denote sufficiently large sets of admissible deformation gradients $\mathbf{F}$ and curl-free electric fields $\mathbf{E}$ consistent with the affine boundary conditions (3).

2.1. Isotropic ideal elastic dielectric composites

As anticipated in the introduction, the object of this paper is to generate solutions for the effective free energy (5) 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 (also referred to in the literature as ideal dielectric elastomers). Namely, the elastic dielectric behaviors of the matrix and particles are characterized by the following free-energy functions

$$W_{\mathbf{m}}(\mathbf{F}, \mathbf{E}) = \begin{cases} \frac{\mu}{2}[I_1 - 3] - \frac{\varepsilon}{2}I_5^E & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \quad \text{and} \quad W_{\mathbf{p}}(\mathbf{F}, \mathbf{E}) = \begin{cases} \frac{\mu_{\mathbf{p}}}{2}[I_1 - 3] - \frac{\varepsilon_{\mathbf{p}}}{2}I_5^E & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases}, \quad (6)$$

where the standard notation $I_1 = \mathbf{F} \cdot \mathbf{F} = F_{ij}F_{ij}$, $I_5^E = \mathbf{F}^{-T} \mathbf{E} \cdot \mathbf{F}^{-T} \mathbf{E}$ has been employed, $\mu, \mu_{\mathbf{p}}$ denote the initial shear moduli of the matrix and the particles, and $\varepsilon, \varepsilon_{\mathbf{p}}$ stand for their respective initial permittivities; basic physical considerations dictate that $\mu, \mu_{\mathbf{p}} > 0$ and $\varepsilon, \varepsilon_{\mathbf{p}} \geq \varepsilon_0$, where ε_0 denotes the permittivity of vacuum. Here, it is appropriate to remark that the choice (6)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 $\mathbf{p} = \mathbf{F}\mathbf{D} - \varepsilon_0 \mathbf{F}^{-T} \mathbf{E}$ is rather insensitive to the state of deformation and varies roughly linearly with the applied Eulerian electric field $\mathbf{e} = \mathbf{F}^{-T} \mathbf{E}$ (see, e.g., Kofod et al., 2003; Wissler and Mazza, 2007; Di Lillo et al., 2011; Cohen et al., 2016). We also note that the choice (6)₂ includes extremal behaviors of notable relevance in applications such as rigid conducting particles, corresponding to the choice $\mu_{\mathbf{p}} = +\infty$ and $\varepsilon_{\mathbf{p}} = +\infty$, and liquid conducting particles, corresponding to $\mu_{\mathbf{p}} = 0$ and $\varepsilon_{\mathbf{p}} = +\infty$ (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 (6), the macroscopic elastic dielectric response of the resulting dielectric elastomer composite is itself isotropic and incompressible. As a result, its effective free-energy function $\bar{W}$ only depends on the macroscopic deformation gradient $\bar{\mathbf{F}}$ and macroscopic Lagrangian electric field $\bar{\mathbf{E}}$ through five invariants and becomes unbounded for non-isochoric deformations when $\bar{J} \doteq \det \bar{\mathbf{F}} \neq 1$. With a slight abuse of notation, we write

$$\bar{W}(\bar{\mathbf{F}}, \bar{\mathbf{E}}, c) = \begin{cases} \bar{W}(\bar{I}_1, \bar{I}_2, \bar{I}_4^E, \bar{I}_5^E, \bar{I}_6^E, c) & \text{if } \bar{J} = 1 \\ +\infty & \text{otherwise} \end{cases} \quad (7)$$

in terms of the five standard invariants

$$\bar{I}_1 = \bar{\mathbf{F}} \cdot \bar{\mathbf{F}}, \quad \bar{I}_2 = \bar{\mathbf{F}}^{-T} \cdot \bar{\mathbf{F}}^{-T}, \quad \bar{I}_4^E = \bar{\mathbf{E}} \cdot \bar{\mathbf{E}}, \quad \bar{I}_5^E = \bar{\mathbf{F}}^{-T} \bar{\mathbf{E}} \cdot \bar{\mathbf{F}}^{-T} \bar{\mathbf{E}}, \quad \bar{I}_6^E = \bar{\mathbf{F}}^{-1} \bar{\mathbf{F}}^{-T} \bar{\mathbf{E}} \cdot \bar{\mathbf{F}}^{-1} \bar{\mathbf{F}}^{-T} \bar{\mathbf{E}}. \quad (8)$$

We shall also find it useful to express $\bar{W}$ alternatively in terms of two of the singular values of $\bar{\mathbf{F}}$, $\bar{\lambda}_1$ and $\bar{\lambda}_2$ say, with the third one $\bar{\lambda}_3 = (\bar{\lambda}_1 \bar{\lambda}_2)^{-1}$, and the three components of the macroscopic Lagrangian electric field, $\bar{E}_1, \bar{E}_2, \bar{E}_3$, with respect to the Lagrangian principal axes (i.e., the principal axes of $\bar{\mathbf{F}}^T \bar{\mathbf{F}}$). With a slight abuse of notation as in (7), we write

$$\bar{W}(\bar{\mathbf{F}}, \bar{\mathbf{E}}, c) = \begin{cases} \bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) & \text{if } \bar{\lambda}_3 = (\bar{\lambda}_1 \bar{\lambda}_2)^{-1} \\ +\infty & \text{otherwise} \end{cases} \quad (9)$$

and note the symmetries $\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = \bar{W}(\bar{\lambda}_2, \bar{\lambda}_1, \bar{E}_2, \bar{E}_1, \bar{E}_3, c) = \bar{W}(\bar{\lambda}_1, (\bar{\lambda}_1 \bar{\lambda}_2)^{-1}, \bar{E}_1, \bar{E}_3, \bar{E}_2, c) = \bar{W}((\bar{\lambda}_1 \bar{\lambda}_2)^{-1}, \bar{\lambda}_1, \bar{E}_3, \bar{E}_1, \bar{E}_2, c) = \bar{W}((\bar{\lambda}_1 \bar{\lambda}_2)^{-1}, \bar{\lambda}_2, \bar{E}_3, \bar{E}_2, \bar{E}_1, c) = \bar{W}(\bar{\lambda}_2, (\bar{\lambda}_1 \bar{\lambda}_2)^{-1}, \bar{E}_2, \bar{E}_3, \bar{E}_1, c)$ and the relations

$$\begin{aligned} \bar{I}_1 &= \bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \frac{1}{\bar{\lambda}_1^2 \bar{\lambda}_2^2}, & \bar{I}_2 &= \frac{1}{\bar{\lambda}_1^2} + \frac{1}{\bar{\lambda}_2^2} + \bar{\lambda}_1^2 \bar{\lambda}_2^2, \\ \bar{I}_4^E &= \bar{E}_1^2 + \bar{E}_2^2 + \bar{E}_3^2, & \bar{I}_5^E &= \frac{\bar{E}_1^2}{\bar{\lambda}_1^2} + \frac{\bar{E}_2^2}{\bar{\lambda}_2^2} + \bar{\lambda}_1^2 \bar{\lambda}_2^2 \bar{E}_3^2, & \bar{I}_6^E &= \frac{\bar{E}_1^2}{\bar{\lambda}_1^4} + \frac{\bar{E}_2^2}{\bar{\lambda}_2^4} + \bar{\lambda}_1^4 \bar{\lambda}_2^4 \bar{E}_3^2. \end{aligned} \quad (10)$$

Power series solution about the ground state $\bar{\mathbf{F}} = \mathbf{I}$ and $\bar{\mathbf{E}} = \mathbf{0}$. For later reference, we note here that the effective free-energy function $\bar{W}$ of (the majority of) isotropic ideal elastic dielectric composites is expected to admit a power series solution about the ground state $\bar{\mathbf{F}} = \mathbf{I}$ and $\bar{\mathbf{E}} = \mathbf{0}$. Expressing such a solution in terms of the variables $\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3$, the finite branch of (9) reads as

$$\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = \sum_{m,n,p,q,r=0}^{\infty} k_{mnpqr} (\bar{\lambda}_1 - 1)^m (\bar{\lambda}_2 - 1)^n \bar{E}_1^p \bar{E}_2^q \bar{E}_3^r, \quad (11)$$

where the coefficients k_{mnpqr} are functions of the microstructure, as characterized by the indicator function θ in (2), as well as of the material parameters $\mu, \mu_p, \varepsilon, \varepsilon_p$.

Given the properties $W_m(\mathbf{F}, -\mathbf{E}) = W_m(\mathbf{F}, \mathbf{E})$ and $W_p(\mathbf{F}, -\mathbf{E}) = W_p(\mathbf{F}, \mathbf{E})$ of the local free energies (6), it follows from the definition (5) of $\bar{W}$ that the electromechanical coupling of the overall response of the composite is even, namely, $\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, -\bar{E}_1, -\bar{E}_2, -\bar{E}_3, c) = \bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c)$. This implies that the coefficients $k_{mnpqr} = 0$ in (11) when $p + q + r = 2n + 1$ with $n \in \mathbb{N}$. Moreover, given the additional properties $W_m(\mathbf{I}, \mathbf{0}) = W_p(\mathbf{I}, \mathbf{0}) = 0$, it follows from the definition (5) that

$$\begin{aligned} k_{00000} &= k_{10000} = k_{01000} = k_{00110} = k_{00101} = k_{00011} = k_{10020} = k_{01200} = 0, \\ k_{20000} &= k_{02000} = k_{11000} = 2\tilde{\mu}, \\ k_{00200} &= k_{00020} = k_{00002} = -\frac{\tilde{\varepsilon}}{2}, \\ k_{10200} &= -k_{10002} = k_{01020} = -k_{01002} = \tilde{m}_K, \end{aligned} \quad (12)$$

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

$$\begin{aligned} \bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) &= 2\tilde{\mu} [(\bar{\lambda}_1 - 1)^2 + (\bar{\lambda}_2 - 1)^2 + (\bar{\lambda}_1 - 1)(\bar{\lambda}_2 - 1)] - \frac{\tilde{\varepsilon}}{2} [\bar{E}_1^2 + \bar{E}_2^2 + \bar{E}_3^2] + \\ &\quad \tilde{m}_K [(\bar{\lambda}_1 - 1)(\bar{E}_1^2 - \bar{E}_3^2) + (\bar{\lambda}_2 - 1)(\bar{E}_2^2 - \bar{E}_3^2)] \end{aligned} \quad (13)$$

plus higher-order correction terms. In the above expressions, $\tilde{\mu}, \tilde{\varepsilon}, \tilde{m}_K$ 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 Tian et al., 2012 and Section II A in Lefèvre and Lopez-Pamies, 2014. In the present context, these effective constants are given by the formulae

$$\begin{aligned} \tilde{\mu} &= \frac{1}{5} \int_{\Omega} \mu(\mathbf{X}) \mathcal{K}_{klmn} \Gamma_{mkl,n} d\mathbf{X}, \\ \tilde{\varepsilon} &= \int_{\Omega} \frac{1}{3} \varepsilon(\mathbf{X}) \gamma_{m,m} d\mathbf{X}, \\ \tilde{m}_K &= \frac{1}{5} \int_{\Omega} \varepsilon(\mathbf{X}) \mathcal{K}_{ijkl} \Gamma_{rij,s} \mathcal{K}_{rspq} \gamma_{p,k} \gamma_{q,l} d\mathbf{X}, \end{aligned} \quad (14)$$

where $\mu(\mathbf{X}) = [1 - \theta(\mathbf{X})] \mu + \theta(\mathbf{X}) \mu_p$, $\varepsilon(\mathbf{X}) = [1 - \theta(\mathbf{X})] \varepsilon + \theta(\mathbf{X}) \varepsilon_p$, $\mathcal{K}_{ijkl} = 1/2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - 1/3\delta_{ij}\delta_{kl}$ with δ_{ij} denoting the Kronecker delta, the notation $_{,i}$ represents partial differentiation with respect to the material point coordinate X_i , and the tensor fields Γ and γ are implicitly defined as the solutions of the following *uncoupled linear* boundary value problems:

$$\begin{cases} [\mu(\mathbf{X}) \mathcal{K}_{ijmn} \Gamma_{mkl,n}]_{,j} = 0 \\ \Gamma_{mkl,m} = 0 \end{cases} \quad \text{for } \mathbf{X} \in \Omega, \quad \Gamma_{ikl} = \delta_{ik} X_l \quad \text{for } \mathbf{X} \in \partial\Omega \quad (15)$$

and

$$[\varepsilon(\mathbf{X}) \gamma_{i,j}]_{,i} = 0 \quad \text{for } \mathbf{X} \in \Omega, \quad \gamma_i = X_i \quad \text{for } \mathbf{X} \in \partial\Omega. \quad (16)$$

3. A solution for an isotropic iterative microstructure

By means of a combination of iterative techniques, Lopez-Pamies (2014) constructed an exact solution for the variational problem (5) for a fairly general class of two-phase particulate microstructures wherein the particles are infinitely polydisperse in size. When specialized to isotropic distributions of filler particles and to matrix and filler particle behaviors characterized by the ideal elastic dielectric free energies (6), his result for the finite branch of the effective free-energy function $\bar{W}$, expressed in terms of the variables $\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3$, can be written as

$$\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = 2\mu\bar{U}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) + \frac{\mu}{2} \left[\bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \frac{1}{\bar{\lambda}_1^2 \bar{\lambda}_2^2} - 3 \right] - \frac{\varepsilon}{2} \left[\frac{\bar{E}_1^2}{\bar{\lambda}_1^2} + \frac{\bar{E}_2^2}{\bar{\lambda}_2^2} + \bar{\lambda}_1^2 \bar{\lambda}_2^2 \bar{E}_3^2 \right]. \quad (17)$$

Here, the function $\bar{U} = \bar{U}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c)$ is defined as the viscosity solution of the first order nonlinear pde

$$c \frac{\partial \bar{U}}{\partial c} - \bar{U} - \sum_{\substack{m,n,p,q,r=0 \\ m+n+p+q+r=2}} \alpha_{mnpqr} \left(\frac{\partial \bar{U}}{\partial \bar{\lambda}_1} \right)^m \left(\frac{\partial \bar{U}}{\partial \bar{\lambda}_2} \right)^n \left(\frac{\partial \bar{U}}{\partial \bar{E}_1} \right)^p \left(\frac{\partial \bar{U}}{\partial \bar{E}_2} \right)^q \left(\frac{\partial \bar{U}}{\partial \bar{E}_3} \right)^r = 0 \quad (18)$$

subject to the initial condition

$$\bar{U}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, 1) = \frac{1}{4} \left(\frac{\mu_p}{\mu} - 1 \right) \left[\bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \frac{1}{\bar{\lambda}_1^2 \bar{\lambda}_2^2} - 3 \right] + \frac{\varepsilon - \varepsilon_p}{4\mu} \left[\frac{\bar{E}_1^2}{\bar{\lambda}_1^2} + \frac{\bar{E}_2^2}{\bar{\lambda}_2^2} + \bar{\lambda}_1^2 \bar{\lambda}_2^2 \bar{E}_3^2 \right], \quad (19)$$

where the fifteen coefficients α_{mnpqr} in (18) are given by expressions (50) in Appendix A due to their bulkiness. The pde (18) 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 $\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3$ 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 (18)–(19) does not appear to admit a closed-form solution. In subsection 3.3, as already announced in the introduction, we present numerical solutions for it that are generated by means of a WENO finite-difference scheme.

3.1. The limit of small deformations and moderate electric fields

Spinelli et al. (2015) recently derived the asymptotic solution for the effective free-energy function of dielectric elastomer composites with the general iterative microstructure of Lopez-Pamies (2014) in the limit of small deformations and moderate electric fields, in the present context, when $\bar{\lambda}_1, \bar{\lambda}_2 \rightarrow 1$ and $\bar{E}_1, \bar{E}_2, \bar{E}_3 \rightarrow 0$. Their solution includes as a special case the solution of interest here for isotropic ideal elastic dielectric composites defined by (17) with (18)–(19). When expressed in terms of the present notation, it reads as

$$\begin{aligned} \bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = & \frac{6(1-c)\mu + 2(2+3c)\mu_p}{(3+2c)\mu + 2(1-c)\mu_p} \mu [(\bar{\lambda}_1 - 1)^2 + (\bar{\lambda}_2 - 1)^2 + (\bar{\lambda}_1 - 1)(\bar{\lambda}_2 - 1)] - \\ & \frac{2(1-c)\varepsilon + (1+2c)\varepsilon_p}{2[(2+c)\varepsilon + (1-c)\varepsilon_p]} \varepsilon [\bar{E}_1^2 + \bar{E}_2^2 + \bar{E}_3^2] + \\ & \left(\varepsilon + \frac{3c(1-c)(\varepsilon_p - \varepsilon)\varepsilon}{10[(2+c)\varepsilon + (1-c)\varepsilon_p]^2} \left(7\varepsilon_p + \frac{(23+7c)\varepsilon}{1-c} - \frac{15\mu(\varepsilon_p - \varepsilon)}{(3+2c)\mu + 2(1-c)\mu_p} \right) \right) \times \\ & [(\bar{\lambda}_1 - 1)(\bar{E}_1^2 - \bar{E}_3^2) + (\bar{\lambda}_2 - 1)(\bar{E}_2^2 - \bar{E}_3^2)] \end{aligned} \quad (20)$$

plus higher-order correction terms. This asymptotic result about the ground state is, of course, of the form (13), where the effective constants are given by

$$\begin{aligned}\tilde{\mu} &= \frac{3(1-c)\mu + (2+3c)\mu_{\text{p}}}{(3+2c)\mu + 2(1-c)\mu_{\text{p}}}\mu, \\ \tilde{\varepsilon} &= \frac{2(1-c)\varepsilon + (1+2c)\varepsilon_{\text{p}}}{(2+c)\varepsilon + (1-c)\varepsilon_{\text{p}}}\varepsilon, \\ \tilde{m}_K &= \varepsilon + \frac{3c(1-c)(\varepsilon_{\text{p}} - \varepsilon)}{10[(2+c)\varepsilon + (1-c)\varepsilon_{\text{p}}]^2} \left(7\varepsilon_{\text{p}} + \frac{(23+7c)\varepsilon}{1-c} - \frac{15\mu(\varepsilon_{\text{p}} - \varepsilon)}{(3+2c)\mu + 2(1-c)\mu_{\text{p}}} \right) \varepsilon.\end{aligned}\quad (21)$$

There are a number of different representations in terms of the set of five standard invariants (8) that are consistent with the asymptotic result (20). For reasons that will become apparent below, here, we spell out one such form:

$$\overline{W}(\overline{I}_1, \overline{I}_2, \overline{I}_4^E, \overline{I}_5^E, \overline{I}_6^E, c) = \frac{\tilde{\mu}}{2} [\overline{I}_1 - 3] + \frac{\tilde{m}_K - \tilde{\varepsilon}}{2} \overline{I}_4^E - \frac{\tilde{m}_K}{2} \overline{I}_5^E. \quad (22)$$

Note that this representation is *linear* in the invariants $\overline{I}_1$, $\overline{I}_4^E$, $\overline{I}_5^E$ and *independent* of the two other invariants $\overline{I}_2$, $\overline{I}_6^E$.

3.2. The limit of infinitely large deformations

In the limit of infinitely large deformations when $\overline{\lambda}_1 \rightarrow 0, +\infty$ and/or $\overline{\lambda}_2 \rightarrow 0, +\infty$, the solution for the effective free-energy function (17) with (18)–(19) can also be worked out in closed form. To avoid loss of continuity, the pertinent asymptotic analysis is summarized in Appendix B. The result can be compactly written in a single expression as

$$\begin{aligned}\overline{W}(\overline{\lambda}_1, \overline{\lambda}_2, \overline{E}_1, \overline{E}_2, \overline{E}_3, c) &= \frac{2(1-c)\mu + (1+2c)\mu_{\text{p}}}{2[(2+c)\mu + 2(1-c)\mu_{\text{p}}]}\mu \left[\overline{\lambda}_1^2 + \overline{\lambda}_2^2 + \frac{1}{\overline{\lambda}_1^2 \overline{\lambda}_2^2} \right] - \\ &\quad \frac{\varepsilon_{\text{p}}}{2[c\varepsilon + (1-c)\varepsilon_{\text{p}}]}\varepsilon \left[\frac{\overline{E}_1^2}{\overline{\lambda}_1^2} + \frac{\overline{E}_2^2}{\overline{\lambda}_2^2} + \overline{\lambda}_1^2 \overline{\lambda}_2^2 \overline{E}_3^2 \right]\end{aligned}\quad (23)$$

plus higher-order correction terms.

An obvious representation in terms of the set of invariants (8) that is consistent with the asymptotic result (23) is given by

$$\overline{W}(\overline{I}_1, \overline{I}_2, \overline{I}_4^E, \overline{I}_5^E, \overline{I}_6^E, c) = \frac{[2(1-c)\mu + (1+2c)\mu_{\text{p}}]}{2[(2+c)\mu + 2(1-c)\mu_{\text{p}}]}\mu \overline{I}_1 - \frac{\varepsilon_{\text{p}}}{2[c\varepsilon + (1-c)\varepsilon_{\text{p}}]}\varepsilon \overline{I}_5^E. \quad (24)$$

Much like the representation (22) for the limit of small deformations and moderate electric fields, the representation (24) for infinitely large deformations is also *linear* in the invariants $\overline{I}_1$, $\overline{I}_4^E$, $\overline{I}_5^E$, with the proportionality constant for $\overline{I}_4^E$ being zero, and *independent* of $\overline{I}_2$, $\overline{I}_6^E$.

3.3. Finite deformations and finite electric fields

For arbitrary values of finite stretches $\overline{\lambda}_1$, $\overline{\lambda}_2$ and finite electric fields $\overline{E}_1$, $\overline{E}_2$, $\overline{E}_3$, the HJ equation (18)–(19) for the function $\overline{U}$ in the effective free energy (17) 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 (Lefèvre et al., 2016). 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 $\overline{\lambda}_1$, $\overline{\lambda}_2$, $\overline{E}_1$, $\overline{E}_2$, $\overline{E}_3$, 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 C.

To gain insight into the qualitative features of the effective free energy (17), we begin by plotting its value, normalized by the initial shear modulus of the matrix material μ , as a function of the stretch $\bar{\lambda}_1$ and the normalized electric field component $\bar{E}_3/\sqrt{\mu/\varepsilon}$ for the experimentally prominent case of axisymmetric electromechanical loading with $\bar{\lambda}_2 = \bar{\lambda}_1$ and $\bar{E}_1 = \bar{E}_2 = 0$. Figure 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. 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 $\varepsilon_p = 10\varepsilon$.

Figure 1: Plots of the effective free energy (17), normalized by the initial shear modulus μ of the underlying matrix material, as a function of the stretch $\bar{\lambda}_1$ and the normalized electric field component $\bar{E}_3/\sqrt{\mu/\varepsilon}$ for axisymmetric electromechanical loading with $\bar{\lambda}_2 = \bar{\lambda}_1$ and $\bar{E}_1 = \bar{E}_2 = 0$. Part (a) corresponds to the case of stiff high-permittivity particles with $\mu_p = 10^2\mu$ and $\varepsilon_p = 10^2\varepsilon$ at volume fraction $c = 0.05$, whereas part (b) corresponds to liquid-like high-permittivity particles with $\mu_p = 10^{-1}\mu$ and $\varepsilon_p = 10\varepsilon$ at volume fraction $c = 0.15$.

Further qualitative as well as quantitative insight into the effective free energy (17) can be gained by plotting its normalized value $\bar{W}/\mu$ as a function of each of the five normalized invariants $\bar{I}_1, \bar{I}_2, \varepsilon\bar{I}_4^E/\mu, \varepsilon\bar{I}_5^E/\mu, \varepsilon\bar{I}_6^E/\mu$ while keeping the remaining four invariants fixed. Figures 2 and 3 show such plots for the same volume fractions of particles and the same two sets of material parameters utilized in Fig. 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 $\bar{I}_1, \bar{I}_2, \varepsilon\bar{I}_4^E/\mu, \varepsilon\bar{I}_5^E/\mu, \varepsilon\bar{I}_6^E/\mu$ restricts the range of physical values that the remaining invariant can take on. For example, for the fixed values $\bar{I}_2 = 8.03, \varepsilon\bar{I}_4^E/\mu = 0.61, \varepsilon\bar{I}_5^E/\mu = 1.63, \varepsilon\bar{I}_6^E/\mu = 10.41$ in Fig. 2(a), the range of physically allowable values of $\bar{I}_1$ is $[5.75, 9.76]$. The results presented in Figs. 2 and 3 span the entire range of physically allowable values for each case that is presented.

Much like in the two preceding asymptotic limits involving small and infinitely large deformations, the key observation from Figs. 2 and 3 is that, for finite deformations and finite electric fields too, the effective free-energy function (17) depends roughly linearly on the invariants $\bar{I}_1, \bar{I}_4^E, \bar{I}_5^E$ and is roughly independent of $\bar{I}_2, \bar{I}_6^E$. The dependence on $\bar{I}_4^E$ is much weaker than on $\bar{I}_1$ and $\bar{I}_5^E$. 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 $\bar{I}_1, \bar{I}_2, \bar{I}_4^E, \bar{I}_5^E, \bar{I}_6^E$ holds true irrespective of the electromechanical properties of the matrix and particles, as measured by $\mu, \mu_p, \varepsilon, \varepsilon_p$, and irrespective of the volume fraction of particles c for the entire range $c \in [0, 1]$ (note that percolation for this class of infinitely polydisperse microstructures occurs at $c = 1$). 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; 2013b), which corresponds to setting $\bar{\mathbf{E}} = \mathbf{0}$ and $\mu_p = +\infty$ in the present context.

Figure 2: Plots of the effective free-energy function (17) of an ideal elastic dielectric, with initial shear modulus μ and initial permittivity ε , 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 $\bar{W}/\mu$ in terms of each of the five normalized invariants $\bar{T}_1$, $\bar{T}_2$, $\varepsilon \bar{T}_4^E/\mu$, $\varepsilon \bar{T}_5^E/\mu$, $\varepsilon \bar{T}_6^E/\mu$ 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 (32).

(a)

(b)

(c)

(d)

(e)

Figure 3: Plots of the effective free-energy function (17) of an ideal elastic dielectric, with initial shear modulus μ and initial permittivity ϵ , filled with liquid-like high-permittivity particles, with initial shear modulus $\mu_p = 10^{-1}\mu$ and initial permittivity $\epsilon_p = 10\epsilon$, at volume fraction $c = 0.15$. Results are shown for the values of the normalized free energy $\bar{W}/\mu$ in terms of each of the five normalized invariants $\bar{I}_1$, $\bar{I}_2$, $\epsilon \bar{I}_4^E/\mu$, $\epsilon \bar{I}_5^E/\mu$, $\epsilon \bar{I}_6^E/\mu$ 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 (32).

4. 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 $\bar{W}$ 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 5 in Part II. Subsection 4.3 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 subsection 4.4.

4.1. 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).

Figure 4: Sample microstructures made out of the periodic repetition of a cubic unit cell with $N = 36$ randomly distributed spherical particles of three different sizes (shown in blue, gray, and red, in order of increasing size) for two particle volume fractions: (a) $c = 0.05$ and (b) $c = 0.15$.

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

$$\{R_p^{(1)}, R_p^{(2)}, R_p^{(3)}\} = \left\{R_p, \frac{7}{9}R_p, \frac{4}{9}R_p\right\} \quad \text{with} \quad R_p = L \left(\frac{3c^{(1)}}{4\pi N_p}\right)^{1/3} \quad (25)$$

and

$$\{c^{(1)}, c^{(2)}, c^{(3)}\} = \{0.5c, 0.25c, 0.25c\} \quad \text{with} \quad c^{(1)} + c^{(2)} + c^{(3)} = c, \quad (26)$$

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 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 $i, j = 1, 2, \dots, N$, must be greater than a certain minimum value s_1 , adjusted by an offset factor $d_1 = 0.02$. This condition reads as

$$||\mathbf{X}^i - \mathbf{X}^j - \mathbf{h}|| \geq s_1, \quad \text{with} \quad s_1 = (R_p^{(m_i)} + R_p^{(m_j)})(1 + d_1), \quad (27)$$

where $\mathbf{X}^i$ ($\mathbf{X}^j$) 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 $\mathbf{h}$ 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

$$|X_k^i - R_p^{(m_i)}| \geq s_2, \quad |X_k^i + R_p^{(m_i)} - L| \geq s_2, \quad s_2 = d_2 R_p^{(m_i)} \quad (k = 1, 2, 3), \quad (28)$$

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

In this work, 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 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.

4.2. 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

$$R_m = L \left(\frac{3c}{4\pi N} \right)^{1/3} \quad (29)$$

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 finite-element discretization):

$$||\mathbf{X}^i - \mathbf{X}^j - \mathbf{h}|| \geq s_1, \quad \text{with} \quad s_1 = 2R_m(1 + d_1) \quad (30)$$

and

$$|X_k^i - R_m| \geq s_2, \quad |X_k^i + R_m - L| \geq s_2, \quad s_2 = d_2 R_m \quad (k = 1, 2, 3) \quad (31)$$

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

Figure 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) $c = 0.15$.

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 5 illustrates two unit cells with $N = 30$ 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.

4.3. The limit of small deformations and moderate electric fields

Figure 6 shows finite-element 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; the interested reader is referred to Appendix A of Spinelli et al. (2015) for details of the finite-element calculations required in this asymptotic limit. 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 (14) with (15)–(16) — in terms of the volume fraction of particles c . Figures 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. 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. 6 for monodisperse particles. To further shed light on this lack of sensitivity to size dispersion, Fig. 6 also includes the analytical result (dotted lines) of Lefèvre and Lopez-Pamies (2014) 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 $c = 0.2$.

Figure 6: Plots of the effective shear modulus $\tilde{\mu}/\mu$, effective permittivity $\tilde{\epsilon}/\epsilon$, and effective electrostrictive constant $\tilde{m}_K/\epsilon$ of a dielectric elastomer composite comprised of an ideal elastic dielectric, with initial shear modulus μ and initial permittivity ϵ , filled with an isotropic distribution of monodisperse spherical particles, with initial shear modulus μ_p and initial permittivity ϵ_p , all as functions of the volume fraction of particles c . Parts (a) and (b) show finite-element results (labeled “Sph. Mono. Exact” and displayed as solid circles) for stiff high-permittivity particles with $\mu_p = 10^2\mu$ and $\epsilon_p = 10^2\epsilon$. Parts (c) and (d) show finite-element results for liquid-like high-permittivity particles with $\mu_p = 10^{-1}\mu$ and $\epsilon_p = 10\epsilon$. The corresponding analytical results of Lefèvre and Lopez-Pamies (2014) 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.

4.4. Finite deformations and finite electric fields

Figures 7 and 8 show finite-element solutions for the effective free-energy function (5) of the ideal elastic dielectric composites with the isotropic distribution of monodisperse spherical particles described in subsection 4.2; again, the technical details of the finite-element calculations are presented in Section 5 of Part II. Similar to the plots shown in Figs. 2 and 3 for the iterative microstructure, with the objective of gaining both qualitative and quantitative insight, Figs. 7 and 8 show plots of the normalized free energy $\bar{W}/\mu$ as a function of each of the five normalized invariants $\bar{I}_1, \bar{I}_2, \bar{\epsilon}_4^E/\mu, \bar{\epsilon}_5^E/\mu, \bar{\epsilon}_6^E/\mu$ for fixed values of the remaining four invariants. The results in Fig. 7 correspond to stiff high-permittivity particles with volume fraction $c = 0.05$ and material parameters $\mu_p = 10^2\mu$ and $\epsilon_p = 10^2\epsilon$, whereas those in Fig. 8 correspond to liquid-like high-permittivity particles with volume fraction $c = 0.15$ and material parameters $\mu_p = 10^{-1}\mu$ and $\epsilon_p = 10\epsilon$. We recall that fixing the values of four of the invariants $\bar{I}_1, \bar{I}_2, \bar{\epsilon}_4^E/\mu, \bar{\epsilon}_5^E/\mu, \bar{\epsilon}_6^E/\mu$ restricts the range of physical values that the remaining invariant can take on. The results shown in Figs. 7 and 8 span the entire range of physically allowable values for each case that is presented.

Figure 7: Plots of the effective free-energy function (5) of a dielectric elastomer composite comprised of an ideal elastic dielectric, with initial shear modulus μ and initial permittivity ε , 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 $\bar{W}/\mu$ in terms of each of the five normalized invariants $\bar{I}_1$, $\bar{I}_2$, $\varepsilon \bar{I}_4^E/\mu$, $\varepsilon \bar{I}_5^E/\mu$, $\varepsilon \bar{I}_6^E/\mu$ for two sets of fixed values of the remaining four invariants. The solid lines (labeled “Sph. Mono. Exact”) correspond to the finite-element solution, while the dashed lines (labeled “Sph. Mono. Approx.”) correspond to the closed-form approximation (32).

Figure 8: Plots of the effective free-energy function (5) of a dielectric elastomer composite comprised of an ideal elastic dielectric, with initial shear modulus μ and initial permittivity ϵ , 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 $\epsilon_p = 10\epsilon$, at volume fraction $c = 0.15$. Results are shown for the values of the normalized free energy $\bar{W}/\mu$ in terms of each of the five normalized invariants $\bar{I}_1$, $\bar{I}_2$, $\epsilon \bar{I}_4^E/\mu$, $\epsilon \bar{I}_5^E/\mu$, $\epsilon \bar{I}_6^E/\mu$ for two sets of fixed values of the remaining four invariants. The solid lines (labeled “Sph. Mono. Exact”) correspond to the finite-element solution, while the dashed lines (labeled “Sph. Mono. Approx.”) correspond to the closed-form approximation (32).

The corresponding results for the isotropic distribution of polydisperse spherical particles described in subsection 4.1 are virtually indistinguishable from those presented in Figs. 7 and 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. 6, but more generally for finite deformations and finite electric fields.

The second and more important observation from the results shown in Figs. 7 and 8 is that the effective free-energy function (5) 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 $\bar{I}_1, \bar{I}_4^E, \bar{I}_5^E$ and is roughly independent of $\bar{I}_2, \bar{I}_6^E$. For this class of microstructures too, the dependence on $\bar{I}_4^E$ is much weaker than on $\bar{I}_1$ and $\bar{I}_5^E$. A broad range of results (in addition to those presented here) have confirmed that such a functional dependence on the invariants $\bar{I}_1, \bar{I}_2, \bar{I}_4^E, \bar{I}_5^E, \bar{I}_6^E$ 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 (at least up to the value of $c = 0.25$ that we managed to consider in our simulations).

5. 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*¹:

$$\bar{W}(\bar{\mathbf{F}}, \bar{\mathbf{E}}, c) = \begin{cases} \frac{\tilde{\mu}}{2} [\bar{I}_1 - 3] + \frac{\tilde{m}_K - \tilde{\varepsilon}}{2} \bar{I}_4^E - \frac{\tilde{m}_K}{2} \bar{I}_5^E & \text{if } \bar{J} = 1 \\ +\infty & \text{otherwise} \end{cases}. \quad (32)$$

Here, we recall that $\bar{I}_1, \bar{I}_4^E, \bar{I}_5^E$ stand for the standard $(\bar{\mathbf{F}}, \bar{\mathbf{E}})$ -based invariants defined by (8)_{1,3,4} and $\tilde{\mu}, \tilde{\varepsilon}, \tilde{m}_K$ denote the effective shear modulus, effective permittivity, and effective electrostrictive constant defined by (14) 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 (15) and (16).

From a practical point of view, we remark that while the second-order linear elliptic pdes (15) and (16) for the tensorial fields Γ and γ 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 $\tilde{\mu}, \tilde{\varepsilon}, \tilde{m}_K$ for any given microstructure. This is why and how. Because of the overall isotropy of the composites of interest here, as already discussed in Lefèvre and Lopez-Pamies (2014), the effective coefficients (14) can be alternatively written as

$$\begin{aligned} \tilde{\mu} &= \int_{\Omega} 2\mu(\mathbf{X}) \mathcal{K}_{12rs} \Gamma_{r12,s} d\mathbf{X}, \\ \tilde{\varepsilon} &= \int_{\Omega} \frac{1}{3} \varepsilon(\mathbf{X}) \gamma_{m,m} d\mathbf{X}, \\ \tilde{m}_K &= \int_{\Omega} 2\varepsilon(\mathbf{X}) \Gamma_{r12,s} \mathcal{K}_{rspq} \gamma_{p,1} \gamma_{q,2} d\mathbf{X}, \end{aligned} \quad (33)$$

¹In addition to the solutions for the three isotropic microstructures discussed heretofore, we have generated finite-element solutions for ideal elastic dielectric composites with other classes of isotropic microstructures, including one wherein the filler particles are of strong anisotropic spheroidal shape. The computed effective free-energy functions (5) for all these different classes of isotropic ideal elastic dielectric composites are accurately described by (32). This appears to indicate that the linearity in $\bar{I}_1, \bar{I}_4^E, \bar{I}_5^E$ and the independence of $\bar{I}_2, \bar{I}_6^E$ of the resulting effective free energies stem from the ideal elastic dielectric nature of the matrix and particles, and not from any particular microstructural trait.

where it is recalled that $\mathcal{K}_{ijkl} = 1/2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - 1/3\delta_{ij}\delta_{kl}$. That is, knowledge of the components $\Gamma_{r12,s}$ and $\gamma_{i,j}$ of the gradients of the fields $\mathbf{\Gamma}$ and $\boldsymbol{\gamma}$ suffices for the computation of the effective constants $\tilde{\mu}$, $\tilde{\varepsilon}$, $\tilde{m}_K$. These components can be directly computed in any commercial FE code that can solve standard *linear* elastostatics and electrostatics problems. Indeed, it follows from (15) that $\Gamma_{i12,j}$ agrees identically with the gradient $u_{i,j}$ of the displacement field u_i solution of the linear elastostatics problem

$$\begin{cases} [\mu(\mathbf{X})(u_{i,j} + u_{j,i})]_{,j} = 0 \\ u_{k,k} = 0 \end{cases} \quad \text{for } \mathbf{X} \in \Omega, \quad u_i = \delta_{i1}X_2 \text{ for } \mathbf{X} \in \partial\Omega. \quad (34)$$

It follows similarly from (16) that $\gamma_{i,j}$ agrees identically with the gradient $\Phi_{,i}$ of the electric potential Φ solution of the linear electrostatics problem

$$[\varepsilon(\mathbf{X})\Phi_{,i}]_{,i} = 0 \quad \text{for } \mathbf{X} \in \Omega, \quad \Phi = X_j \quad j = 1, 2, 3 \text{ for } \mathbf{X} \in \partial\Omega. \quad (35)$$

Having generated numerical solutions for $\Gamma_{r12,s}$ and $\gamma_{i,j}$, the integrals (33) 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 $\tilde{m}_K$.

The macroscopic constitutive relation (4) implied by the effective free-energy function (32) is given by

$$\bar{\mathbf{S}} = \tilde{\mu}\bar{\mathbf{F}} + \tilde{m}_K\bar{\mathbf{F}}^{-T}\bar{\mathbf{E}} \otimes \bar{\mathbf{F}}^{-1}\bar{\mathbf{F}}^{-T}\bar{\mathbf{E}} - \bar{p}\bar{\mathbf{F}}^{-T}, \quad (36)$$

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

$$\bar{\mathbf{D}} = (\tilde{\varepsilon} - \tilde{m}_K)\bar{\mathbf{E}} + \tilde{m}_K\bar{\mathbf{F}}^{-1}\bar{\mathbf{F}}^{-T}\bar{\mathbf{E}}. \quad (37)$$

By construction, the effective free energy (32) is exact in the limit of small deformations and moderate electric fields as $\bar{\mathbf{F}} \rightarrow \mathbf{I}$ and $\bar{\mathbf{E}} \rightarrow \mathbf{0}$. Also by construction, while not exact, the effective free energy (32) is expected to be extremely accurate for finite deformations and finite electric fields given that it is linear in $\bar{I}_1$, $\bar{I}_4^E$, $\bar{I}_5^E$ and independent of $\bar{I}_2$, $\bar{I}_6^E$. This expectation is supported by the direct comparisons shown above in Figs. 2, 3, 7, 8 and further below in Figs. 10, 11 with the exact solutions for the three considered microstructures. Indeed, the results based on (32), 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 μ , μ_p , ε , ε_p , as well as for all choices of volume fractions of particles c .

5.1. The $\bar{\mathbf{F}}$ and $\bar{\mathbf{D}}$ formulation

Depending on the specific problem at hand, it may be more convenient to utilize the macroscopic electric displacement field $\bar{\mathbf{D}}$ as the independent electric variable instead of the macroscopic electric field $\bar{\mathbf{E}}$ utilized in (32). This can be readily accomplished with help of the following partial Legendre transform:

$$\begin{aligned} \bar{W}^*(\bar{\mathbf{F}}, \bar{\mathbf{D}}, c) &= \sup_{\bar{\mathbf{E}}} \{ \bar{\mathbf{D}} \cdot \bar{\mathbf{E}} + \bar{W}(\bar{\mathbf{F}}, \bar{\mathbf{E}}, c) \} \\ &= \begin{cases} \frac{\tilde{\mu}}{2} [\bar{I}_1 - 3] + \frac{1}{2\tilde{m}_K} \left[\frac{\bar{I}_5^D + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^2 \bar{I}_4^D + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) [\bar{I}_1 \bar{I}_5^D - \bar{I}_6^D]}{1 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^3 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^2 \bar{I}_2 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{I}_1} \right] & \text{if } \bar{J} = 1 \\ +\infty & \text{otherwise} \end{cases}, \end{aligned} \quad (38)$$

where the invariants $\bar{I}_1$, $\bar{I}_2$ are given, again, by expressions (8)_{1,2}, and

$$\bar{I}_4^D = \bar{\mathbf{D}} \cdot \bar{\mathbf{D}}, \quad \bar{I}_5^D = \bar{\mathbf{F}}\bar{\mathbf{D}} \cdot \bar{\mathbf{F}}\bar{\mathbf{D}}, \quad \bar{I}_6^D = \bar{\mathbf{F}}^T\bar{\mathbf{F}}\bar{\mathbf{D}} \cdot \bar{\mathbf{F}}^T\bar{\mathbf{F}}\bar{\mathbf{D}}. \quad (39)$$

Physically, the potential $\bar{W}^*$ defined by (38) corresponds to the macroscopic Helmholtz free energy of the composite. It follows that the first Piola-Kirchhoff stress tensor $\bar{\mathbf{S}}$ and electric field $\bar{\mathbf{E}}$ can be written in terms of the deformation gradient $\bar{\mathbf{F}}$ and electric displacement $\bar{\mathbf{D}}$ simply as

$$\begin{aligned}\bar{\mathbf{S}} &= \frac{\partial \bar{W}^*}{\partial \bar{\mathbf{F}}}(\bar{\mathbf{F}}, \bar{\mathbf{D}}, c) - \bar{q} \bar{\mathbf{F}}^{-T} \\ &= \tilde{\mu} \bar{\mathbf{F}} - \bar{q} \bar{\mathbf{F}}^{-T} + \frac{1}{\tilde{m}_K \left[1 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^3 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^2 \bar{I}_2 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{I}_1 \right]} \times \\ &\quad \left[\left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \left[\bar{I}_5^D \bar{\mathbf{F}} - \bar{\mathbf{F}} \bar{\mathbf{D}} \otimes \bar{\mathbf{F}}^T \bar{\mathbf{F}} \bar{\mathbf{D}} - \bar{\mathbf{F}} \bar{\mathbf{F}}^T \bar{\mathbf{F}} \bar{\mathbf{D}} \otimes \bar{\mathbf{D}} \right] + \left(1 + \bar{I}_1 \frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{\mathbf{F}} \bar{\mathbf{D}} \otimes \bar{\mathbf{D}} \right] - \\ &\quad \frac{\left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{I}_5^D + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^3 \bar{I}_4^D + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^2 [\bar{I}_1 \bar{I}_5^D - \bar{I}_6^D]}{\tilde{m}_K \left[1 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^3 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^2 \bar{I}_2 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{I}_1 \right]^2} \left[\bar{\mathbf{F}} - \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{\mathbf{F}}^{-T} \bar{\mathbf{F}}^{-1} \bar{\mathbf{F}}^{-T} \right], \quad (40)\end{aligned}$$

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

$$\begin{aligned}\bar{\mathbf{E}} &= \frac{\partial \bar{W}^*}{\partial \bar{\mathbf{D}}}(\bar{\mathbf{F}}, \bar{\mathbf{D}}, c) \\ &= \frac{1}{\tilde{m}_K \left[1 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^3 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^2 \bar{I}_2 + \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{I}_1 \right]} \left[\left(1 + \bar{I}_1 \frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{\mathbf{F}}^T \bar{\mathbf{F}} \bar{\mathbf{D}} + \right. \\ &\quad \left. \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right)^2 \bar{\mathbf{D}} - \left(\frac{\tilde{\varepsilon} - \tilde{m}_K}{\tilde{m}_K} \right) \bar{\mathbf{F}}^T \bar{\mathbf{F}} \bar{\mathbf{F}}^T \bar{\mathbf{F}} \bar{\mathbf{D}} \right]. \quad (41)\end{aligned}$$

We note that the (finite branch of the) effective free energy (38) in terms of $\bar{\mathbf{F}}$ and $\bar{\mathbf{D}}$ is *not* of the separable form $\bar{W}^* = \bar{W}_{elas}^*(\bar{I}_1, \bar{I}_2, c) + \bar{W}_{elec}^*(\bar{I}_4^D, \bar{I}_5^D, \bar{I}_6^D, c)$, 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 (32) of the effective free energy written in terms of $\bar{\mathbf{F}}$ and $\bar{\mathbf{E}}$, which is of the separable form $\bar{W} = \bar{W}_{elas}(\bar{I}_1, \bar{I}_2, c) + \bar{W}_{elec}(\bar{I}_4^E, \bar{I}_5^E, \bar{I}_6^E, c)$. We further note that the effective free energy (38) *does* depend on all five invariants $\bar{I}_1, \bar{I}_2, \bar{I}_4^D, \bar{I}_5^D, \bar{I}_6^D$, whereas (32) is independent of $\bar{I}_2$ and $\bar{I}_6^E$.

5.2. Material instabilities

In addition to facilitating the computation of the macroscopic electromechanical constitutive response (40)–(41), the free-energy function (38) in terms of $\bar{\mathbf{F}}$ and $\bar{\mathbf{D}}$ 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. 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)

$$\min_{\substack{\bar{\mathbf{F}}, \bar{\mathbf{D}} \\ \text{tr}(\bar{\mathbf{F}} \bar{\mathbf{F}}^{-1})=0}} \left\{ \dot{F}_{ij} \bar{\mathcal{L}}_{ijkl}(\bar{\mathbf{F}}, \bar{\mathbf{D}}, c) \dot{F}_{kl} + 2 \dot{F}_{ij} \bar{\mathcal{M}}_{ijk}(\bar{\mathbf{F}}, \bar{\mathbf{D}}, c) \dot{D}_k + \dot{D}_i \bar{\mathcal{B}}_{ij}(\bar{\mathbf{F}}, \bar{\mathbf{D}}, c) \dot{D}_j \right\} > 0. \quad (42)$$

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)

$$\min_{\substack{\mathbf{u}, \mathbf{v} \\ \|\mathbf{u}\|=\|\mathbf{v}\|=1, \mathbf{u} \cdot \mathbf{v}=0}} \left\{ \mathbf{v} \cdot \left[\hat{\mathbf{K}} - \frac{2}{(\text{tr} \hat{\mathbf{B}})^2 - \text{tr} \hat{\mathbf{B}}^2} \hat{\mathbf{R}} \left(\text{tr}(\hat{\mathbf{B}}) \hat{\mathbf{I}} - \hat{\mathbf{B}} \right) \hat{\mathbf{R}}^T \right] \mathbf{v} \right\} > 0, \quad (43)$$

where $\widehat{\mathbf{K}} = \widehat{\mathbf{I}}\mathbf{K}\widehat{\mathbf{I}}$, $\widehat{\mathbf{R}} = \widehat{\mathbf{I}}\mathbf{R}\widehat{\mathbf{I}}$, $\widehat{\mathbf{B}} = \widehat{\mathbf{I}}\mathbf{B}\widehat{\mathbf{I}}$ with $K_{ik} = \overline{F}_{ja}\overline{F}_{lb}\overline{\mathcal{L}}_{iakb}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c)u_ju_l$, $R_{ik} = \overline{F}_{ja}\overline{F}_{bk}^{-1}\overline{\mathcal{M}}_{iab}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c)u_j$, $B_{ij} = \overline{F}_{ai}^{-1}\overline{F}_{bj}^{-1}\overline{\mathcal{B}}_{ab}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c)$, and $\hat{I}_{ij} = \delta_{ij} - u_iu_j$. In the above expressions,

$$\overline{\mathcal{L}}_{ijkl}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c) = \frac{\partial^2 \overline{W}^*}{\partial \overline{F}_{ij} \partial \overline{F}_{kl}}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c), \quad \overline{\mathcal{M}}_{ijk}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c) = \frac{\partial^2 \overline{W}^*}{\partial \overline{F}_{ij} \partial \overline{D}_k}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c), \quad \overline{\mathcal{B}}_{ij}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c) = \frac{\partial^2 \overline{W}^*}{\partial \overline{D}_i \partial \overline{D}_j}(\overline{\mathbf{F}}, \overline{\mathbf{D}}, c), \quad (44)$$

where, with a slight abuse of notation, $\overline{W}^*$ in these derivatives stands for the finite branch of the effective free energy (38).

The sets of all critical points $(\overline{\mathbf{F}}_{cr}, \overline{\mathbf{D}}_{cr})$ at which conditions (42) and (43) first fail along a continuous isochoric loading path, with starting point the ground state $(\overline{\mathbf{F}}, \overline{\mathbf{D}}) = (\mathbf{I}, \mathbf{0})$, 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 work to study these failure surfaces in their entirety, but we do study the possible failure of conditions (42) and (43) for electromechanical boundary conditions encountered in typical electrostriction experiments, which are described next.

5.3. Electrostriction

One of the archetype experiments to probe the performance of dielectric elastomers consists in exposing them to an uniaxial electric field and measuring the resulting deformation, commonly referred to as electrostriction. In practice, as shown schematically in Fig. 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 (32) 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 Rossi, 2005; McCarthy et al., 2009; Meddeb and Ouanies, 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.

Figure 9: Schematic of the typical experimental setup, in (left) the undeformed and (right) the deformed configurations, to probe the electrostriction of dielectric elastomers under the application of an uniaxial Lagrangian electric field $\overline{\mathbf{E}} = -\Phi/L_3$.

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

$$\overline{\mathbf{S}}_{ij} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \overline{\mathbf{E}}_i = \begin{bmatrix} 0 \\ 0 \\ \overline{E} \end{bmatrix}. \quad (45)$$

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. 9. It follows from the constitutive relations (36) and (37) that

$$\bar{F}_{ij} = \begin{bmatrix} \bar{\lambda}^{-1/2} & 0 & 0 \\ 0 & \bar{\lambda}^{-1/2} & 0 \\ 0 & 0 & \bar{\lambda} \end{bmatrix} \quad \text{and} \quad \bar{D}_i = \begin{bmatrix} 0 \\ 0 \\ \bar{D} \end{bmatrix}, \quad (46)$$

where the electrostriction stretch $\bar{\lambda}$ in the direction of the applied electric field (see Fig. 9) and the non-trivial component $\bar{D}$ of the electric displacement are defined by the relations

$$\bar{\lambda}^4 - \bar{\lambda} + \frac{\tilde{m}_K}{\tilde{\mu}} \bar{E}^2 = 0 \quad \text{and} \quad \bar{D} = \left[\tilde{\varepsilon} - \tilde{m}_K \left(1 - \frac{1}{\bar{\lambda}^2} \right) \right] \bar{E} \quad (47)$$

in terms of the applied electric field $\bar{E}$.

In the absence of filler particles when $c = 0$, $\tilde{\mu} = \mu$ and $\tilde{\varepsilon} = \tilde{m}_K = \varepsilon$, and hence the constitutive relations (47) reduce of course to those of the unfilled elastic dielectric matrix: $\bar{\lambda}^4 - \bar{\lambda} + \varepsilon \bar{E}^2 / \mu = 0$ and $\bar{D} = \varepsilon \bar{E} / \bar{\lambda}^2$. By comparing these to the general form of (47) 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 $\tilde{m}_K / \tilde{\mu}$ and the values of $\tilde{\varepsilon}$ and $\tilde{m}_K$. Sample numerical results are provided below in Figs. 10 and 11 for two cases of practical relevance.

Material instabilities. Along an electromechanical loading path of the form (45) where $|\bar{E}|$ is continuously increased from the ground state $\bar{E} = 0$, condition (42) first fails when

$$|\bar{E}| = \sqrt{\frac{3\tilde{\mu}}{2^{8/3}\tilde{m}_K}} \doteq \bar{E}_{LPD}. \quad (48)$$

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

$$\bar{\lambda}_{LPD} = 2^{-2/3} \quad \text{and} \quad \bar{D}_{LPD} = \left| \tilde{\varepsilon} + \left(2^{4/3} - 1 \right) \tilde{m}_K \right| \sqrt{\frac{3\tilde{\mu}}{2^{8/3}\tilde{m}_K}}. \quad (49)$$

We can readily deduce from (48) that for microstructures and particle behaviors for which $\tilde{\mu} / \tilde{m}_K < \mu / \varepsilon$, the addition of filler particles shifts the limiting electric field $\bar{E}_{LPD}$ to smaller values than that of the matrix $\sqrt{3\mu / 2^{8/3}\varepsilon}$. By contrast, for microstructures and particle behaviors for which $\tilde{\mu} / \tilde{m}_K > \mu / \varepsilon$, the addition of filler particles delays the attainment of the electromechanical limit load. Further, in view of (49)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 (49)₂, much like the critical electric field (48), does depend on the ratio $\tilde{\mu} / \tilde{m}_K$ as well as on the values of $\tilde{\varepsilon}$ 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 $\bar{\lambda}_{LPD}$ and $\bar{E}_{LPD}$ are presented in Figs. 10 and 11.

As opposed to condition (42) and irrespectively of the material parameters of the matrix and particles and also of the content of particles, the strong ellipticity condition (43) never fails under conditions of electrostriction (45).

Sample results. Figures 10 and 11 present sample results determined from the proposed approximate effective free energy (32) — labeled “Aprox.” and displayed as dashed lines — for the macroscopic response and stability of an ideal elastic dielectric (with material parameters μ and ε), filled with the infinitely polydisperse and monodisperse² isotropic distributions of ideal elastic dielectric particles (with material

²Again, 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. 10 and 11 for monodisperse spherical particles can be viewed as corresponding to isotropic distributions of polydisperse spherical particles as well.

parameters μ_p , ε_p , and volume fraction c) described in Sections 3 and 4, under the conditions of electrostriction (45). In particular, Fig. 10 presents results for stiff high-permittivity particles with material parameters $\mu_p = 10^2$ and $\varepsilon_p = 10^2\varepsilon$, whereas Fig. 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 (32), the numerical viscosity solutions for the infinitely polydisperse iterative microstructure and the finite-element 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. 10(a) through (d) show plots of the electrostriction stretch $\bar{\lambda}$ in terms of the applied electric field $\bar{E}$, normalized by the quantity $\sqrt{\mu/\varepsilon}$, as characterized by equation (47)₁. Figures 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. 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 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 of Lefèvre and Lopez-Pamies (2014) and Spinelli et al. (2015) 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 Part II.

Figures 10(a) through (d) also clearly illustrate the attainment of a maximum electric field at the critical stretch $\bar{\lambda}_{LPD} = 2^{-2/3}$, 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. 9, electrostriction stretches $\bar{\lambda} < \bar{\lambda}_{LPD}$ would indeed be accessible if instead of a voltage, charges were applied on the compliant electrodes. The value of the maximum electric field $\bar{E}_{LPD}$ given by relation (48), which does depend on the specifics of the microstructure, is plotted in Fig. 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 $\bar{E}_{LPD}$ with the addition of particles. On the other hand, the composite with spherical particles exhibits modestly smaller values of $\bar{E}_{LPD}$ as the content of particles is increased.

Figures 11(a) through (d) show results analogous to those shown in Figs. 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. This is accompanied, however, by a significant reduction in the limiting electric field $\bar{E}_{LPD}$. Finally, a quick glance at Fig. 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.

Figure 10: Electrostriction response and stability determined from the effective free energy (32) under conditions (45) — labeled “Approx.” and displayed as dashed lines in the plots — for an ideal elastic dielectric of initial shear modulus μ and initial permittivity ε 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 (32), corresponding plots are also included of the numerical viscosity solution for the iterative microstructure (labeled “Exact” and displayed as solid lines) and of the finite-element solutions for the microstructure with spherical particles (labeled “Exact” and displayed as solid lines or solid circles).

Figure 11: Electrostriction response and stability determined from the effective free energy (32) under conditions (45) — labeled “Approx.” and displayed as dashed lines in the plots — for an ideal elastic dielectric of initial shear modulus μ and initial permittivity ε filled with liquid-like high-permittivity particles of initial shear modulus $\mu_p = 10^{-1}\mu$, initial permittivity $\varepsilon_p = 10\varepsilon$, 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 (32), corresponding plots are also included of the numerical viscosity solution for the iterative microstructure (labeled “Exact” and displayed as solid lines) and of the finite-element solutions for the microstructure with spherical particles (labeled “Exact” and displayed as solid lines or solid circles).

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. 11 certainly motivate more experimental studies of the latter.

6. Final comments

The present work 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 (32) 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:

$$\overline{W}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, c_p, c_i) = \min_{\mathbf{F} \in \mathcal{K}} \max_{\mathbf{E} \in \mathcal{E}} \int_{\Omega} W(\mathbf{X}, \mathbf{F}, \mathbf{E}) d\mathbf{X}, \quad W(\mathbf{X}, \mathbf{F}, \mathbf{E}) = \begin{cases} \frac{\mu(\mathbf{X})}{2}[I_1 - 3] - \frac{\varepsilon(\mathbf{X})}{2}I_5^E & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases}$$

with

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

and

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

Here, θ_p and θ_i denote the indicator functions of the spatial regions occupied in the ground state Ω 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 $\overline{I}_1$, $\overline{I}_4^E$, $\overline{I}_5^E$ and are independent of $\overline{I}_2$, $\overline{I}_6^E$, irrespectively of the specifics of their heterogeneity.

By way of an example, in support of the above conjecture, Fig. 12(a) illustrates the close agreement between the result (dashed line) determined from the approximation (32) and from a finite-element solution (solid line) for the $\overline{\lambda}$ vs. $\overline{E}/\sqrt{\mu/\varepsilon}$ electrostriction response of a *three-phase* composite made out of an ideal elastic dielectric matrix, with material parameters μ and ε , 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 (14) are given by $\tilde{\mu} = 1.1583\mu$, $\tilde{\varepsilon} = 1.2044\varepsilon$, $\tilde{m}_K = 1.1783\varepsilon$. Figure 12(b) illustrates the unit cell whose periodic repetition defines the precise microstructure in this example.

Figure 12: (a) Comparison of the electrostriction response (see subsection 5.3 for the definition of the pertinent variables) determined from the approximate closed-form free energy (32) and from a finite-element 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 μ and initial permittivity ε) 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 ω , and material parameters μ_p , ε_p of the particles are indicated in the figure, as so are the volume fraction c_i and material parameters μ_i , ε_i of the interphases.

Acknowledgements

Support for this work by the National Science Foundation through the CAREER Grant CMMI-1219336 (formerly CMMI-1055528) is gratefully acknowledged.

Appendix A. The coefficients α_{mnpqr}

The fifteen coefficients α_{mnpqr} in the pde (18) read as follows:

$$\begin{aligned}
\alpha_{20000} &= \frac{2\Gamma_E \bar{\lambda}_1 \bar{\lambda}_2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^8 - 2\bar{\lambda}_1^6 \bar{\lambda}_2^6 + 2\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)^2} - \frac{\Gamma_F \bar{\lambda}_1^5 \bar{\lambda}_2^3 \left(3\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 2\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right) \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)^2} - \frac{\bar{\lambda}_1^4 \bar{\lambda}_2^6 - 2\bar{\lambda}_1^6 \bar{\lambda}_2^4 + \bar{\lambda}_1^2 \bar{\lambda}_2^2 - \bar{\lambda}_1^8 \bar{\lambda}_2^2 + \bar{\lambda}_1^4}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)}, \\
\alpha_{02000} &= \frac{2\Gamma_E \bar{\lambda}_1 \bar{\lambda}_2^3 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 2\bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1 \right)}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} - \frac{\Gamma_F \bar{\lambda}_1^3 \bar{\lambda}_2^5}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right) \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} - \frac{\bar{\lambda}_1^6 \bar{\lambda}_2^4 - 2\bar{\lambda}_1^4 \bar{\lambda}_2^6 + \bar{\lambda}_1^2 \bar{\lambda}_2^2 - \bar{\lambda}_1^2 \bar{\lambda}_2^8 + \bar{\lambda}_2^4}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)}, \\
\alpha_{11000} &= \frac{2\Gamma_E \bar{\lambda}_1^2 \bar{\lambda}_2^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 + \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 2 \right)}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} + \frac{2\Gamma_F \bar{\lambda}_1^4 \bar{\lambda}_2^4}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right) \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} - \frac{2\bar{\lambda}_1 \bar{\lambda}_2 \left(\bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)}{3 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^2}, \\
\alpha_{00200} &= \frac{\Gamma_E \bar{\lambda}_1^3 \bar{\lambda}_2 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)}{\left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right) \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} \left[\frac{2\bar{E}_1^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^8 - 2\bar{\lambda}_1^6 \bar{\lambda}_2^6 + 2\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)}{3\bar{\lambda}_1^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right) \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right) \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)} + \frac{\bar{E}_2^2 \left(\bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 + \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 2 \right)}{3\bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^3 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)} - \right. \\
&\quad \left. \frac{\bar{E}_3^2 \bar{\lambda}_1^4 \bar{\lambda}_2^4 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 2\bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1 \right)}{3 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)^3 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)} - \frac{\mu}{\varepsilon} \right] + \frac{\Gamma_F \bar{\lambda}_1^5 \bar{\lambda}_2^3}{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1} \left[\frac{\bar{E}_1^2 \left(2\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 3\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1 \right)}{3\bar{\lambda}_1^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right) \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)} + \right. \\
&\quad \left. \frac{\bar{E}_2^2 \left(\bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)^2}{3\bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^3} + \frac{2\bar{E}_3^2 \bar{\lambda}_1^4 \bar{\lambda}_2^4 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1 \right)^2}{3 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)^3} - \frac{\mu}{\varepsilon} \right] + \frac{\bar{\lambda}_1^2 \bar{\lambda}_2^2}{\bar{\lambda}_1^2 - \bar{\lambda}_2^2} \left[\frac{\bar{E}_1^2 \left(\bar{\lambda}_1^6 \bar{\lambda}_2^2 + 2\bar{\lambda}_1^4 \bar{\lambda}_2^4 - \bar{\lambda}_1^2 \bar{\lambda}_2^6 - \bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)}{3\bar{\lambda}_1^2 \bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right) \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} - \right. \\
&\quad \left. \frac{4\bar{E}_2^2 \bar{\lambda}_1^2 \left(\bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)}{3\bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^3} + \frac{\bar{E}_3^2 \bar{\lambda}_1^{10} \bar{\lambda}_2^{10} \left(\bar{\lambda}_1^8 \bar{\lambda}_2^4 - 2\bar{\lambda}_1^6 \bar{\lambda}_2^6 - 2\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)}{3\bar{\lambda}_1^4 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)^3} + \frac{\mu}{\varepsilon} \right], \\
\alpha_{00020} &= \frac{\Gamma_E \bar{\lambda}_1 \bar{\lambda}_2^3}{\bar{\lambda}_1^2 - \bar{\lambda}_2^2} \left[\frac{\bar{E}_1^2 \left(\bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 + \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 2 \right)}{3\bar{\lambda}_1^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^3 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} + \frac{\bar{E}_3^2 \bar{\lambda}_1^4 \bar{\lambda}_2^4 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 2\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1 \right) \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1 \right)^2}{3 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right) \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)^3} + \right. \\
&\quad \left. \frac{2\bar{E}_2^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 2\bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1 \right)}{3\bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right) \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)} + \frac{\mu}{\varepsilon} \right] + \frac{\Gamma_F \bar{\lambda}_1^3 \bar{\lambda}_2^3}{3 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)} \left[\frac{\bar{E}_1^2 \bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)^2 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)}{\bar{\lambda}_1^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^3 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} - \frac{\bar{E}_2^2}{\bar{\lambda}_1^2 - \bar{\lambda}_2^2} + \right. \\
&\quad \left. \frac{\bar{E}_3^2 \bar{\lambda}_1^4 \bar{\lambda}_2^6 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1 \right)^2}{\left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)^2 \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} \right] - \frac{\bar{\lambda}_1^2 \bar{\lambda}_2^2}{\bar{\lambda}_1^2 - \bar{\lambda}_2^2} \left[\frac{4\bar{E}_1^2 \bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)}{3\bar{\lambda}_1^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right)^3} + \frac{\bar{E}_2^2 \left(\bar{\lambda}_1^6 \bar{\lambda}_2^2 - 2\bar{\lambda}_1^4 \bar{\lambda}_2^4 - \bar{\lambda}_1^2 \bar{\lambda}_2^6 + \bar{\lambda}_1^2 + \bar{\lambda}_2^2 \right)}{3\bar{\lambda}_1^2 \bar{\lambda}_2^2 \left(\bar{\lambda}_1^2 - \bar{\lambda}_2^2 \right) \left(\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1 \right)} - \right. \\
&\quad \left. \frac{\bar{E}_3^2 \bar{\lambda}_1^2 \bar{\lambda}_2^2 \left(2\bar{\lambda}_1^6 \bar{\lambda}_2^6 - \bar{\lambda}_1^4 \bar{\lambda}_2^8 + 2\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1 \right)}{3 \left(\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1 \right)^3} + \frac{\mu}{\varepsilon} \right],
\end{aligned}$$

$$\begin{aligned}
\alpha_{00002} &= \frac{\Gamma_E \bar{\lambda}_1 \bar{\lambda}_2}{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1} \left[\frac{\bar{E}_1^2 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1)^2 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 2 \bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1)}{3 \bar{\lambda}_1^2 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1)^3} + \frac{\bar{E}_2^2 (\bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1)^2 (2 \bar{\lambda}_1^4 \bar{\lambda}_2^2 - \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)}{3 \bar{\lambda}_2^2 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)^3} - \right. \\
&\quad \left. \frac{2 \bar{E}_3 \bar{\lambda}_1 \bar{\lambda}_2^4 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 + \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 2)}{3 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1) (\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)} + \frac{\mu}{\varepsilon} \right] + \frac{\Gamma_F \bar{\lambda}_1 \bar{\lambda}_2}{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1} \left[\frac{2 \bar{E}_1^2 \bar{\lambda}_2^2 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1)^2}{3 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1)^3} + \frac{\bar{E}_2^2 \bar{\lambda}_1^2 (\bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1)^2}{3 (\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)^3} - \right. \\
&\quad \left. \frac{\bar{E}_3^2 \bar{\lambda}_1 \bar{\lambda}_2^4 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 + 2 \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 3)}{3 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1) (\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)} + \frac{\mu}{\varepsilon} \right] + \frac{\bar{E}_1^2 (\bar{\lambda}_1^8 \bar{\lambda}_2^4 - 2 \bar{\lambda}_1^6 \bar{\lambda}_2^6 - 2 \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)}{3 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1)^3} + \\
&\quad \frac{\bar{E}_2^2 (2 \bar{\lambda}_1^6 \bar{\lambda}_2^6 - \bar{\lambda}_1^4 \bar{\lambda}_2^8 + 2 \bar{\lambda}_1^4 \bar{\lambda}_2^2 + 1)}{3 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)^3} - \frac{\bar{E}_3^2 \bar{\lambda}_1^2 \bar{\lambda}_2^2 (\bar{\lambda}_1^6 \bar{\lambda}_2^2 - \bar{\lambda}_1^2 \bar{\lambda}_2^6 - \bar{\lambda}_1^2 + \bar{\lambda}_2^2)}{3 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1) (\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)}, \\
\alpha_{00110} &= -\frac{8 \bar{E}_1 \bar{E}_2}{3 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2)^3} \left[\frac{\Gamma_E \bar{\lambda}_1^3 \bar{\lambda}_2^3 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 + \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 2)}{(\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1)} + \frac{\Gamma_F \bar{\lambda}_1^5 \bar{\lambda}_2^5}{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1} + \frac{(\bar{\lambda}_1^2 + \bar{\lambda}_2^2) (\bar{\lambda}_1^4 - 6 \bar{\lambda}_1^2 \bar{\lambda}_2^2 + \bar{\lambda}_2^4)}{4 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2)} \right], \\
\alpha_{00011} &= \frac{8 \bar{E}_2 \bar{E}_3 \bar{\lambda}_1^2 \bar{\lambda}_2^2}{3 (\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)^3} \left[\frac{\Gamma_E \bar{\lambda}_1^3 \bar{\lambda}_2^5 (2 \bar{\lambda}_1^4 \bar{\lambda}_2^2 - \bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1)}{(\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1)} - \frac{\Gamma_F \bar{\lambda}_1^5 \bar{\lambda}_2^7}{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1} - \frac{\bar{\lambda}_1^6 \bar{\lambda}_2^8 - \bar{\lambda}_1^4 \bar{\lambda}_2^{10} + 4 \bar{\lambda}_1^4 \bar{\lambda}_2^4 - \bar{\lambda}_1^2 + \bar{\lambda}_2^2}{4 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2)} \right], \\
\alpha_{00101} &= \frac{8 \bar{E}_1 \bar{E}_3 \bar{\lambda}_1^2 \bar{\lambda}_2^2}{3 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1)^3} \left[\frac{\Gamma_E \bar{\lambda}_1^5 \bar{\lambda}_2^3 (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 2 \bar{\lambda}_1^2 \bar{\lambda}_2^4 + 1)}{(\bar{\lambda}_1^2 - \bar{\lambda}_2^2) (\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1)} - \frac{2 \Gamma_F \bar{\lambda}_1^7 \bar{\lambda}_2^5}{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1} - \frac{\bar{\lambda}_1^{10} \bar{\lambda}_2^4 - \bar{\lambda}_1^8 \bar{\lambda}_2^6 - 4 \bar{\lambda}_1^4 \bar{\lambda}_2^4 - \bar{\lambda}_1^2 + \bar{\lambda}_2^2}{4 (\bar{\lambda}_1^2 - \bar{\lambda}_2^2)} \right], \\
\alpha_{10100} &= \frac{2 \bar{E}_1}{\bar{\lambda}_1} \alpha_{20000}, \quad \alpha_{10010} = \frac{\bar{E}_2}{\bar{\lambda}_2} \alpha_{11000}, \quad \alpha_{10001} = -\frac{2 \bar{E}_3}{\bar{\lambda}_1} \alpha_{20000} - \frac{\bar{E}_3}{\bar{\lambda}_2} \alpha_{11000}, \\
\alpha_{01100} &= \frac{\bar{E}_1}{\bar{\lambda}_1} \alpha_{11000}, \quad \alpha_{01010} = \frac{2 \bar{E}_2}{\bar{\lambda}_2} \alpha_{02000}, \quad \alpha_{01001} = -\frac{2 \bar{E}_3}{\bar{\lambda}_2} \alpha_{02000} - \frac{\bar{E}_3}{\bar{\lambda}_1} \alpha_{11000}. \tag{50}
\end{aligned}$$

In the above expressions,

$$\begin{aligned}
\Gamma_F &= \frac{1}{\sqrt{1 - \bar{\lambda}_1^2 \bar{\lambda}_2^4}} \mathcal{E}_F \left\{ \frac{\sqrt{\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1}}{2 \sqrt{1 - \bar{\lambda}_1^2 \bar{\lambda}_2^4}} \ln \left[2 \bar{\lambda}_1 \bar{\lambda}_2^2 \left(\sqrt{\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1} + \bar{\lambda}_1 \bar{\lambda}_2^2 \right) - 1 \right]; \frac{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1}{\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1} \right\}, \\
\Gamma_E &= \frac{1}{\sqrt{1 - \bar{\lambda}_1^2 \bar{\lambda}_2^4}} \mathcal{E}_E \left\{ \frac{\sqrt{\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1}}{2 \sqrt{1 - \bar{\lambda}_1^2 \bar{\lambda}_2^4}} \ln \left[2 \bar{\lambda}_1 \bar{\lambda}_2^2 \left(\sqrt{\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1} + \bar{\lambda}_1 \bar{\lambda}_2^2 \right) - 1 \right]; \frac{\bar{\lambda}_1^4 \bar{\lambda}_2^2 - 1}{\bar{\lambda}_1^2 \bar{\lambda}_2^4 - 1} \right\}, \tag{51}
\end{aligned}$$

where the functions $\mathcal{E}_F$ and $\mathcal{E}_E$, stand for, respectively, the incomplete elliptic integrals of first and second kind:

$$\mathcal{E}_F \{\phi; \nu\} = \int_0^\phi [1 - \nu^2 \sin^2 \theta]^{-1/2} d\theta, \quad \mathcal{E}_E \{\phi; \nu\} = \int_0^\phi [1 - \nu^2 \sin^2 \theta]^{1/2} d\theta. \tag{52}$$

Appendix B. Asymptotic analysis of the Hamilton-Jacobi equation (18)–(19) in the limit of infinitely large deformations

This appendix outlines the derivation of the asymptotic solution (23) for the effective free-energy function (17) with (18)–(19) in the limit of infinitely large deformations when $\bar{\lambda}_1 \rightarrow 0, +\infty$ and/or $\bar{\lambda}_2 \rightarrow 0, +\infty$.

Numerical solutions of the HJ equation (18)–(19) for $\bar{U} = \bar{U}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c)$ suggest that

$$\bar{U}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = \frac{2f(c) - \mu}{4\mu} \left[\bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \frac{1}{\bar{\lambda}_1^2 \bar{\lambda}_2^2} \right] + \frac{2g(c) + \varepsilon}{4\mu} \left[\frac{\bar{E}_1^2}{\bar{\lambda}_1^2} + \frac{\bar{E}_2^2}{\bar{\lambda}_2^2} + \bar{\lambda}_1^2 \bar{\lambda}_2^2 \bar{E}_3^2 \right] + HOT \quad (53)$$

and hence that

$$\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = f(c) \left[\bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \frac{1}{\bar{\lambda}_1^2 \bar{\lambda}_2^2} \right] + g(c) \left[\frac{\bar{E}_1^2}{\bar{\lambda}_1^2} + \frac{\bar{E}_2^2}{\bar{\lambda}_2^2} + \bar{\lambda}_1^2 \bar{\lambda}_2^2 \bar{E}_3^2 \right] + HOT \quad (54)$$

for large and small values of $\bar{\lambda}_1$ and/or $\bar{\lambda}_2$, where f and g are functions of the volume fraction of particles c . Substituting the ansatz (53) in the HJ equation (18)–(19) 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 $\bar{W}$ presented next.

In the limit as $\bar{\lambda}_1 \rightarrow +\infty$, $\bar{W}$ takes on the form

$$\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = \frac{2(1-c)\mu + (1+2c)\mu_p}{2[(2+c)\mu + 2(1-c)\mu_p]} \mu \bar{\lambda}_1^2 - \frac{\varepsilon_p}{2[c\varepsilon + (1-c)\varepsilon_p]} \varepsilon \bar{\lambda}_1^2 \bar{\lambda}_2^2 \bar{E}_3^2 + o(\bar{\lambda}_1^2). \quad (55)$$

In the opposite limit when $\bar{\lambda}_1 \rightarrow 0$, $\bar{W}$ reads as

$$\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = \frac{2(1-c)\mu + (1+2c)\mu_p}{2[(2+c)\mu + 2(1-c)\mu_p]} \mu \frac{1}{\bar{\lambda}_1^2 \bar{\lambda}_2^2} - \frac{\varepsilon_p}{2[c\varepsilon + (1-c)\varepsilon_p]} \varepsilon \frac{\bar{E}_1^2}{\bar{\lambda}_1^2} + o(\bar{\lambda}_1^{-2}). \quad (56)$$

It follows from the symmetry condition $\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3) = \bar{W}(\bar{\lambda}_2, \bar{\lambda}_1, \bar{E}_2, \bar{E}_1, \bar{E}_3)$ that $\bar{W}$ is given by

$$\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = \frac{2(1-c)\mu + (1+2c)\mu_p}{2[(2+c)\mu + 2(1-c)\mu_p]} \mu \bar{\lambda}_2^2 - \frac{\varepsilon_p}{2[c\varepsilon + (1-c)\varepsilon_p]} \varepsilon \bar{\lambda}_1^2 \bar{\lambda}_2^2 \bar{E}_3^2 + o(\bar{\lambda}_2^2), \quad (57)$$

when $\bar{\lambda}_2 \rightarrow +\infty$ and by

$$\bar{W}(\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3, c) = \frac{2(1-c)\mu + (1+2c)\mu_p}{2[(2+c)\mu + 2(1-c)\mu_p]} \mu \frac{1}{\bar{\lambda}_1^2 \bar{\lambda}_2^2} - \frac{\varepsilon_p}{2[c\varepsilon + (1-c)\varepsilon_p]} \varepsilon \frac{\bar{E}_2^2}{\bar{\lambda}_2^2} + o(\bar{\lambda}_2^{-2}), \quad (58)$$

when $\bar{\lambda}_2 \rightarrow 0$. The four different asymptotic expressions (55)–(58) can be gathered into the single asymptotic expression (23) presented in the main body of the text for general loading.

Appendix C. Numerical viscosity solution of the Hamilton-Jacobi equation (18)–(19)

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 (18)–(19) for the function $\bar{U}$ in the effective free energy (17); the derivation and complete description of the scheme will be reported elsewhere (Lefèvre et al., 2016). For simplicity of exposition, we present the scheme in the context of axisymmetric electromechanical loading conditions with

$$\bar{\lambda}_1 = \bar{\lambda}_2, \quad \bar{E}_1 = \bar{E}_2 = 0 \quad \text{and employ the notation} \quad \bar{\lambda}_1 = x, \quad \bar{E}_3 = y. \quad (59)$$

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 (18)–(19) involving all five space variables $\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3$ shall be apparent.

We begin by defining the function $u(x, y, c) = \bar{U}(x, x, 0, 0, y, c)$ and note that

$$\frac{\partial \bar{U}}{\partial x}(x, x, 0, 0, y, c) = \frac{\partial \bar{U}}{\partial y}(x, x, 0, 0, y, c) = \frac{1}{2} \frac{\partial u}{\partial x}(x, y, c). \quad (60)$$

Under the axisymmetric conditions of loading (59), it follows that the HJ equation (18)–(19) for $\bar{U}$ reduces to the simpler HJ equation for u :

$$\frac{\partial u}{\partial c} + \mathcal{H}\left(x, y, c, u, \frac{\partial u}{\partial x}, \frac{\partial u}{\partial y}\right) = 0, \quad u(x, y, 1) = \frac{1}{4} \left(\frac{\mu_p}{\mu} - 1 \right) \left[2x^2 + \frac{1}{x^4} - 3 \right] + \frac{\varepsilon - \varepsilon_p}{4\mu} x^4 y^2, \quad (61)$$

where the Hamiltonian $\mathcal{H}$ in the pde (61)₁ is given by

$$\mathcal{H}(x, y, c, u, p, q) = -\frac{u}{c} - \frac{\beta_1(x)}{c} p^2 - \left(\frac{4y^2 \beta_1(x)}{cx^2} + \frac{\mu \beta_2(x)}{\varepsilon c x^4} \right) q^2 + \frac{4y \beta_1(x)}{cx} pq \quad (62)$$

with

$$\beta_1(x) = \frac{(2x^6 + 1)x^6}{12(x^6 - 1)^2} - \frac{x^{12} \ln[\sqrt{x^{-6} - 1} + x^{-3}]}{4(1 - x^6)^{5/2}} \quad \text{and} \quad \beta_2(x) = \frac{x^6}{1 - x^6} - \frac{\ln[\sqrt{x^{-6} - 1} + x^{-3}]}{(x^{-6} - 1)^{3/2} x^3}. \quad (63)$$

Monotone numerical Hamiltonian

We seek to compute the viscosity solution of (61) with Hamiltonian given by (62). 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 (61) 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 $u_{x,i,j}^+ = \Delta_x^+ u_{k,l}/h_x = (u_{k+1,l} - u_{k,l})/h_x$, $u_{x,i,j}^- = \Delta_x^- u_{k,l}/h_x = (u_{k,l} - u_{k-1,l})/h_x$, $u_{y,i,j}^+ = \Delta_y^+ u_{k,l}/h_y = (u_{k,l+1} - u_{k,l})/h_y$, $u_{y,i,j}^- = \Delta_y^- u_{k,l}/h_y = (u_{k,l} - u_{k,l-1})/h_y$, first-order monotone schemes refer to schemes of the form

$$\frac{d}{dc} u_{i,j} = -\widehat{\mathcal{H}}(x_i, y_j, c, u_{i,j}, u_{x,i,j}^+, u_{x,i,j}^-, u_{y,i,j}^+, u_{y,i,j}^-), \quad (64)$$

where $\widehat{\mathcal{H}}$, the so-called numerical Hamiltonian (also termed flux), is a Lipschitz continuous function such that it is *consistent* with the Hamiltonian $\mathcal{H}$ in the sense that

$$\widehat{\mathcal{H}}(x, y, c, u, p, p, q, q) = \mathcal{H}(x, y, c, u, p, q), \quad (65)$$

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,

$$\widehat{\mathcal{H}}(x, y, c, u, \downarrow, \uparrow, \downarrow, \uparrow). \quad (66)$$

There are a number of monotone numerical Hamiltonians that have been proposed in the literature. In this work, we make use of the so-called Roe flux with LLF entropy correction (Osher and Shu, 1991). Omitting the dependence on x, y, c , and u to ease notation, the Roe flux with LLF entropy correction reads as:

$$\widehat{\mathcal{H}}(p^+, p^-, q^+, q^-) = \begin{cases} \mathcal{H}(p^*, q^*) & \text{if } \mathcal{H}_1(p, q) \text{ and } \mathcal{H}_2(p, q) \text{ do not change} \\ & \text{signs in } p \in I(p^-, p^+), q \in I(q^-, q^+); \\ \mathcal{H}\left(\frac{p^+ + p^-}{2}, q^*\right) - \nu_1(p^+, p^-) \frac{p^+ - p^-}{2} & \text{otherwise and if } \mathcal{H}_2(p, q) \text{ does not} \\ & \text{change sign in } A \leq p \leq B, q \in I(q^-, q^+); \\ \mathcal{H}\left(p^*, \frac{q^+ + q^-}{2}\right) - \nu_2(q^+, q^-) \frac{q^+ - q^-}{2} & \text{otherwise and if } \mathcal{H}_1(p, q) \text{ does not} \\ & \text{change sign in } p \in I(p^-, p^+), C \leq q \leq D; \\ \widehat{\mathcal{H}}^{LLF}(p^+, p^-, q^+, q^-) & \text{otherwise} \end{cases} \quad (67)$$

where p^* and q^* are defined by

$$p^* = \begin{cases} p^+ & \text{if } \mathcal{H}_1(p, q) \leq 0 \\ p^- & \text{if } \mathcal{H}_1(p, q) \geq 0 \end{cases}, \quad q^* = \begin{cases} q^+ & \text{if } \mathcal{H}_2(p, q) \leq 0 \\ q^- & \text{if } \mathcal{H}_2(p, q) \geq 0 \end{cases}, \quad (68)$$

ν_1 and ν_2 are defined by

$$\nu_1(p^+, p^-) = \max_{\substack{p \in I(p^-, p^+) \\ C \leq q \leq D}} |\mathcal{H}_1(p, q)|, \quad \nu_2(q^+, q^-) = \max_{\substack{q \in I(q^-, q^+) \\ A \leq p \leq B}} |\mathcal{H}_2(p, q)|, \quad (69)$$

and

$$\widehat{\mathcal{H}}^{LLF}(p^+, p^-, q^+, q^-) = \mathcal{H}\left(\frac{p^+ + p^-}{2}, \frac{q^+ + q^-}{2}\right) - \nu_1(p^+, p^-) \frac{p^+ - p^-}{2} - \nu_2(q^+, q^-) \frac{q^+ - q^-}{2}. \quad (70)$$

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 $\widehat{\mathcal{H}}$, 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, Osher, and Chan (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).

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 $\{(x_i, y_j) : x_0 < x_1 < \dots < x_{m-1} < x_m, \ y_0 < y_1 < \dots < y_{n-1} < y_n \text{ with } x_{i+1} - x_i = h_x = h, \ y_{j+1} - y_j = h_y = h \ \forall (i, j) \in \{0, 1, 2, \dots, m\} \times \{0, 1, 2, \dots, n\}\}$, 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 (67) is given by the formula

$$u_{x,i,j}^- = \frac{1}{12} \left(-\frac{\Delta_x^+ u_{i-2,j}}{h} + 7\frac{\Delta_x^+ u_{i-1,j}}{h} + 7\frac{\Delta_x^+ u_{i,j}}{h} - \frac{\Delta_x^+ u_{i+1,j}}{h} \right) - g \left(\frac{\Delta_x^- \Delta_x^+ u_{i-2,j}}{h}, \frac{\Delta_x^- \Delta_x^+ u_{i-1,j}}{h}, \frac{\Delta_x^- \Delta_x^+ u_{i,j}}{h}, \frac{\Delta_x^- \Delta_x^+ u_{i+1,j}}{h} \right). \quad (71)$$

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

$$u_{x,i,j}^+ = \frac{1}{12} \left(-\frac{\Delta_x^+ u_{i-2,j}}{h} + 7\frac{\Delta_x^+ u_{i-1,j}}{h} + 7\frac{\Delta_x^+ u_{i,j}}{h} - \frac{\Delta_x^+ u_{i+1,j}}{h} \right) + g \left(\frac{\Delta_x^- \Delta_x^+ u_{i+2,j}}{h}, \frac{\Delta_x^- \Delta_x^+ u_{i+1,j}}{h}, \frac{\Delta_x^- \Delta_x^+ u_{i,j}}{h}, \frac{\Delta_x^- \Delta_x^+ u_{i-1,j}}{h} \right). \quad (72)$$

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

$$u_{y,i,j}^- = \frac{1}{12} \left(-\frac{\Delta_y^+ u_{i-2,j}}{h} + 7\frac{\Delta_y^+ u_{i-1,j}}{h} + 7\frac{\Delta_y^+ u_{i,j}}{h} - \frac{\Delta_y^+ u_{i+1,j}}{h} \right) - g \left(\frac{\Delta_y^- \Delta_y^+ u_{i-2,j}}{h}, \frac{\Delta_y^- \Delta_y^+ u_{i-1,j}}{h}, \frac{\Delta_y^- \Delta_y^+ u_{i,j}}{h}, \frac{\Delta_y^- \Delta_y^+ u_{i+1,j}}{h} \right). \quad (73)$$

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

$$u_{y,i,j}^+ = \frac{1}{12} \left(-\frac{\Delta_y^+ u_{i,j-2}}{h} + 7\frac{\Delta_y^+ u_{i,j-1}}{h} + 7\frac{\Delta_y^+ u_{i,j}}{h} - \frac{\Delta_y^+ u_{i,j+1}}{h} \right) + g \left(\frac{\Delta_y^- \Delta_y^+ u_{i,j+2}}{h}, \frac{\Delta_y^- \Delta_y^+ u_{i,j+1}}{h}, \frac{\Delta_y^- \Delta_y^+ u_{i,j}}{h}, \frac{\Delta_y^- \Delta_y^+ u_{i,j-1}}{h} \right). \quad (74)$$

In expressions (71) through (74),

$$g(z_1, z_2, z_3, z_4) = \frac{1}{3} \hat{\omega}^{(1)} (z_1 - 2z_2 + z_3) + \frac{1}{6} (\hat{\omega}^{(3)} - \frac{1}{2}) (z_2 - 2z_3 + z_4) \quad (75)$$

with

$$\hat{\omega}^{(l)} = \frac{\frac{\gamma^{(l)}}{(\epsilon + h^2 \hat{\mathcal{S}}^{(l)})^2}}{\sum_{k=1}^3 \frac{\gamma^{(k)}}{(\epsilon + h^2 \hat{\mathcal{S}}^{(k)})^2}}, \quad \gamma^{(1)} = \frac{1}{10}, \quad \gamma^{(2)} = \frac{3}{5}, \quad \gamma^{(3)} = \frac{3}{10}, \quad \epsilon = 10^{-6}, \quad (76)$$

and

$$\hat{\mathcal{S}}^{(k)} = \frac{13}{12} (z_k - z_{k+1})^2 + [(k-2)z_k - (k-3)z_{k+1}]^2, \quad (77)$$

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 (71)–(74) 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 by Lefèvre et al. (2016) which maintain the fifth-order accuracy of the scheme. For conciseness, we do not report such expressions here and refer the interested reader to Lefèvre et al. (2016).

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 (64) in the time variable $\mathbf{c}$. 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 $\mathbf{c}^n < \mathbf{c}^{n-1} < \dots < \mathbf{c}^2 < \mathbf{c}^1 < \mathbf{c}^0$ with $\mathbf{c}^{k+1} - \mathbf{c}^k = \Delta \mathbf{c}$ for all $k = \{0, 1, 2, \dots, n\}$, where $\Delta \mathbf{c}$ is a constant. For grid sizes $h_x = h_y = h$ and $\Delta \mathbf{c}$, we denote by $u_{i,j}^n$ a numerical approximation to the viscosity solution of (64) at the space point $(x, y) = (x_i, y_j)$ and time $\mathbf{c} = \mathbf{c}^n$, namely, $u(x_i, y_j, \mathbf{c}^n) = u(ih, jh, 1 + n\Delta \mathbf{c})$. Given $u_{i,j}^n$, the algorithm to compute $u_{i,j}^{n+1}$ is as follows. Let

$$L_{i,j}^{(l)} \{ \mathbf{c} \} = -\hat{\mathcal{H}}(x_i, y_j, \mathbf{c}, u_{i,j}^{(l)}, u_{x,i,j}^{(l)+}, u_{x,i,j}^{(l)-}, u_{y,i,j}^{(l)+}, u_{y,i,j}^{(l)-}), \quad (78)$$

where, again, the numerical Hamiltonian $\hat{\mathcal{H}}$ is given by the Roe flux (67) 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 (71)–(74) 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:

$$\begin{aligned}
u_{i,j}^{(1)} &= u_{i,j}^n, & k_{i,j}^{(1)} &= L_{i,j}^{(1)} \{c^n\}, \\
u_{i,j}^{(2)} &= u_{i,j}^{(1)} + \frac{1}{2} \Delta c k_{i,j}^{(1)}, & k_{i,j}^{(2)} &= L_{i,j}^{(2)} \left\{ c^n + \frac{1}{2} \Delta c \right\}, \\
u_{i,j}^{(3)} &= u_{i,j}^{(1)} + \frac{1}{16} \Delta c (3k_{i,j}^{(1)} + k_{i,j}^{(2)}), & k_{i,j}^{(3)} &= L_{i,j}^{(3)} \left\{ c^n + \frac{1}{4} \Delta c \right\} \\
u_{i,j}^{(4)} &= u_{i,j}^{(1)} + \frac{1}{2} \Delta c k_{i,j}^{(3)}, & k_{i,j}^{(4)} &= L_{i,j}^{(4)} \left\{ c^n + \frac{1}{2} \Delta c \right\}, \\
u_{i,j}^{(5)} &= u_{i,j}^{(1)} + \frac{3}{16} \Delta c (-k_{i,j}^{(2)} + 2k_{i,j}^{(3)} + 3k_{i,j}^{(4)}), & k_{i,j}^{(5)} &= L_{i,j}^{(5)} \left\{ c^n + \frac{3}{4} \Delta c \right\}, \\
u_{i,j}^{(6)} &= u_{i,j}^{(1)} + \frac{1}{7} \Delta c (k_{i,j}^{(1)} + 4k_{i,j}^{(2)} + 6k_{i,j}^{(3)} - 12k_{i,j}^{(4)} + 8k_{i,j}^{(5)}), & k_{i,j}^{(6)} &= L_{i,j}^{(6)} \{c^n + \Delta c\}, \\
u_{i,j}^{n+1} &= u_{i,j}^n + \frac{\Delta c}{90} \left(7k_{i,j}^{(1)} + 32k_{i,j}^{(3)} + 12k_{i,j}^{(4)} + 32k_{i,j}^{(5)} + 7k_{i,j}^{(6)} \right). \tag{79}
\end{aligned}$$

The corresponding formulae for the general HJ equation (18)–(19) involving all five space variables $\bar{\lambda}_1, \bar{\lambda}_2, \bar{E}_1, \bar{E}_2, \bar{E}_3$ should be now apparent.

References

- [1] Benton, S.H., 1977. *The Hamilton-Jacobi Equation: A Global Approach*. Academic Press, New York.
- [2] Carpi, F., De Rossi, D., 2005. Improvement of electromechanical actuating performances of a silicone dielectric elastomer by dispersion of titanium dioxide powder. *IEEE Transactions on Dielectrics and Electrical Insulation* 12, 835–843.
- [3] Cohen, N., Dayal, K., deBotton, G., 2016. Electroelasticity of polymer networks. *J. Mech. Phys. Solids* 92, 105–126.
- [4] Crandall, M., Lions, P., 1983. Viscosity solutions of Hamilton-Jacobi equations. *Trans. Amer. Math. Soc.* 277, 1–42.
- [5] Crandall, M., Lions, P., 1984. Two approximations of solutions of Hamilton-Jacobi equations. *Math. Comput.* 43, 1–19.
- [6] Dang, Z.-M., Fan, L.-Z., Shen, Y., Nan, C.-W., 2003. Study on dielectric behavior of a three-phase CF/(PVDF + BaTiO₃) composite. *Chemical Physics Letters* 369, 95–100.
- [7] Di Lillo, L., Schmidt, A., Bergamini, A., Ermanni, P., Mazza, E., 2011. Dielectric and insulating properties of an acrylic DEA material at high near-DC electric fields. *Proc. of SPIE* 7976, 79763B.
- [8] Dorfmann, A., Ogden, R.W., 2005. Nonlinear electroelasticity. *Acta Mechanica* 174, 167–183.
- [9] Fassler, A., Majidi, C., 2015. Liquid-phase metal inclusions for a conductive polymer composite. *Adv. Mater.* 27, 1928–1932.
- [10] Geymonat, G., Müller, S., Triantafyllidis, N., 1993. Homogenization of nonlinearly elastic materials, microscopic bifurcation and macroscopic loss of rank-one convexity. *Archive for Rational Mechanics and Analysis* 122, 231–290.
- [11] Goudarzi, T., Spring, D.W., Paulino, G.H., Lopez-Pamies, O., 2015. Filled elastomers: A theory of filler reinforcement based on hydrodynamic and interphasial effects. *Journal of the Mechanics and Physics of Solids* 80, 37–67.
- [12] Gusev, A.A., 1997. Representative volume element size for elastic composites: a numerical study. *J. Mech. Phys. Solids* 45, 1449–1459.
- [13] Hill, R., 1957. On uniqueness and stability in the theory of finite elastic strain. *J. Mech. Phys. Solids* 5, 229–241.
- [14] Hill, R., 1972. On constitutive macrovariables for heterogeneous solids at finite strain. *Proc. R. Soc. Lond. A* 326, 131–147.
- [15] Huang, C., Zhang, Q.M., 2004. Enhanced dielectric and electromechanical response in high-dielectric constant all-polymer percolative composites. *Advanced Functional Materials* 14, 501–506.
- [16] Huang, C., Zhang, Q.M., Li, J.Y., Rabeony, M., 2005. Colossal dielectric and electromechanical responses in self-assembled polymeric nanocomposites. *Applied Physics Letters* 87, 182901.
- [17] Jiang, G.-S., Shu, C.-W., 1996. Efficient implementation of weighted ENO schemes. *J. Comput. Phys.* 126, 202–228.
- [18] Kofod, G., Sommer-Larsen, P., Kornbluh, R., Pelrine, R., 2003. Actuation response of polyacrylate dielectric elastomers. *J. Intell. Mater. Syst. Struct.* 14, 787–793.
- [19] Lawson, J.D., 1966. An order five Runge-Kutta process with extended region of stability. *SIAM J. Numer. Anal.* 3, 593–597.
- [20] Leblanc, J.L., 2010. *Filled Polymers: Science and Industrial Applications*. CRC Press, Boca Raton.
- [21] Lefèvre, V., Lopez-Pamies, O., 2014. The overall elastic dielectric properties of a suspension of spherical particles in rubber: An exact explicit solution in the small-deformation limit. *Journal of Applied Physics* 116, 134106.
- [22] Lefèvre, V., Lopez-Pamies, O., 2016. Nonlinear electroelastic deformations of dielectric elastomer composites: II — Non-Gaussian elastic dielectrics. Submitted.
- [23] Lefèvre, V., Garnica, A., Lopez-Pamies, O., 2016. A WENO finite-difference scheme for a new class of Hamilton-Jacobi equations in nonlinear solid mechanics. In preparation.
- [24] Lewis, T.J., 2004. Interfaces are the dominant feature of dielectrics at the nanometric level. *IEEE Transactions on Dielectrics and Electrical Insulation* 11, 739–753.

- [25] Li, J.Y., 2003. Exchange coupling in P(VDF-TrFE) copolymer based all-organic composites with giant electrostriction. *Phys. Rev. Lett.* 90, 217601–217604.
- [26] Li, J.Y., Rao, N., 2004. Micromechanics of ferroelectric polymer-based electrostrictive composites. *J. Mech. Phys. Solids* 52, 591–615.
- [27] Li, J.Y., Huang, C., Zhang, Q., 2004. Enhanced electromechanical properties in all-polymer percolative composites. *Appl. Phys. Lett.* 84, 3124.
- [28] Liu, X.-D., Osher, S., Chan, T., 1994. Weighted essentially non-oscillatory schemes. *J. Comput. Phys.* 115, 200–212.
- [29] Liu, H., Zhang, L., Yang, D., Yu, Y., Yao, L., Tian, M., 2013. Mechanical, dielectric, and actuated strain of silicone elastomer filled with various types of TiO_2 . *Soft Materials* 11, 363–370.
- [30] Lopez-Pamies, O., 2014. Elastic dielectric composites: Theory and application to particle-filled ideal dielectrics. *J. Mech. Phys. Solids* 64, 61–82.
- [31] Lopez-Pamies, O., Goudarzi, T., Nakamura, T., 2013a. The nonlinear elastic response of suspensions of rigid inclusions in rubber: I — An exact result for dilute suspensions. *J. Mech. Phys. Solids* 61, 1–18.
- [32] Lopez-Pamies, O., Goudarzi, T., Danas, K., 2013b. The nonlinear elastic response of suspensions of rigid inclusions in rubber: II — A simple explicit approximation for finite-concentration suspensions. *J. Mech. Phys. Solids* 61, 19–37.
- [33] Lopez-Pamies, O., Goudarzi, T., Meddeb, A.B., Ounaies, Z., 2014. Extreme enhancement and reduction of the dielectric response of polymer nanoparticulate composites via interphasial charges. *Applied Physics Letters* 104, 242904.
- [34] McCarthy, D.N., Risse, S., Katekomol, P., Kofod, G., 2009. The effect of dispersion on the increased relative permittivity of TiO_2 /SEBS composites. *J. Phys. D: Appl. Phys.* 42, 145406.
- [35] Meddeb, A.M., Ounaies, Z., 2012. Nano-enhanced polymer composites for energy storage applications. *Proc. SPIE* 8342, 834207.
- [36] Michel, J.C., Moulinec, H., Suquet, P., 1999. Effective properties of composite materials with periodic microstructure: a computational approach. *Comput. Methods Appl. Mech. Eng.* 172, 109–143.
- [37] Miehe, C. Vallicotti, D., Teichtmeister, S., 2016. Homogenization and multiscale stability analysis in finite magneto-electro-elasticity. Application to soft matter EE, ME and MEE composites. *Comput. Methods Appl. Mech. Engrg.* 300, 294–346.
- [38] Nan, C.-W., Cai, N., Liu, L., Zhai, J., Ye, Y., Lin, Y., 2003. Coupled magnetic-electric properties and critical behavior in multiferroic particulate composites. *J. Appl. Phys.* 94, 5930–5936.
- [39] Osher, S., Sethian, J., 1988. Fronts propagating with curvature dependent speed: algorithms based on Hamilton-Jacobi formulations. *J. Comput. Phys.* 79, 12–49.
- [40] Osher, S., Shu, C.-W., 1991. High-order essentially nonoscillatory schemes for Hamilton-Jacobi equations. *SIAM J. Numer. Anal.* 28, 907–922.
- [41] Peltine, R., Kornbluh, R., Joseph, J.P., 1998. Electrostriction of polymer dielectrics with compliant electrodes as a means of actuation. *Sensors and Actuators A* 64, 77–85.
- [42] Ponte Castañeda, P., Siboni, M., 2012. A finite-strain constitutive theory for electro-active polymer composites via homogenization. *International Journal of Non Linear Mechanics* 47, 293–306.
- [43] Segurado, J., Llorca, J., 2002. A numerical approximation to the elastic properties of sphere-reinforced composites. *J. Mech. Phys. Solids* 50, 2107–2121.
- [44] Shu, C.-W., 2009. High order weighted essentially nonoscillatory schemes for convection dominated problems. *SIAM Review* 51, 82–126.
- [45] Siboni, M.H., Ponte Castañeda, P., 2013. Dielectric elastomer composites: small-deformation theory and applications. *Philosophical Magazine* 93, 2769–2801.
- [46] Spinelli, S.A., Lopez-Pamies, O., 2015. Some simple explicit results for the elastic dielectric properties and stability of layered composites. *International Journal of Engineering Science* 88, 15–28.
- [47] Spinelli, S.A., Lefèvre, V., Lopez-Pamies, O., 2015. Dielectric elastomer composites: A general closed-form solution in the small-deformation limit. *J. Mech. Phys. Solids* 83, 263–284.
- [48] Stratton, J.S., 1941. *Electromagnetic Theory*. McGraw-Hill.
- [49] Tian, L., 2007. *Effective Behavior of Dielectric Elastomer Composites*. Ph.D. Thesis, California Institute of Technology.
- [50] Tian, L., Tevet-Deree, L., deBotton, G., Bhattacharya, K., 2012. Dielectric elastomer composites. *J. Mech. Phys. Solids* 60, 181–198.
- [51] Wissler, M., Mazza, E., 2007. Electromechanical coupling in dielectric elastomer actuators. *Sensors and Actuators A* 138, 384–393.
- [52] Zhang, Q.M., Hengfeng, L., Poh, M., Xia, F., Cheng, Z.-Y., Xu, H., Huang, C., 2002. An all-organic composite actuator material with high dielectric constant. *Nature* 419, 284–287.
- [53] Zhao, X., Suo, Z., 2007. Method to analyze electromechanical stability of dielectric elastomers. *Appl. Phys. Lett.* 91, 061921.