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Elastic dielectric composites: Theory and application to particle-filled ideal dielectrics

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ABSTRACT

A microscopic field theory is developed with the aim of describing, explaining, and predicting the macroscopic response of elastic dielectric composites with two-phase particulate (periodic or random) microstructures under arbitrarily large deformations and electric fields. The central idea rests on the construction — via an iterated homogenization technique in finite electroelastostatics — of a specific but yet fairly general class of particulate microstructures which allow to compute exactly the homogenized response of the resulting composite materials. The theory is applicable to any choice of elastic dielectric behaviors (with possibly even or odd electroelastic coupling) for the underlying matrix and particles, and any choice of the one- and two-point correlation functions describing the microstructure. In spite of accounting for fine microscopic information, the required calculations amount to solving tractable first-order nonlinear (Hamilton-Jacobi-type) partial differential equations.

As a first application of the theory, explicit results are worked out for the basic case of ideal elastic dielectrics filled with initially spherical particles that are distributed either isotropically or in chain-like formations and that are ideal elastic dielectrics themselves. The effects that the permittivity, stiffness, volume fraction, and spatial distribution of the particles have on the overall electrostrictive deformation (induced by the application of a uniaxial electric field) of the composite are discussed in detail.

1. Introduction

Following their discovery in the 19th century (see, e.g., the historical review by Cady, 1946), deformable dielectrics have progressively enabled a wide variety of technologies. This has been particularly true for “hard” deformable dielectrics such as piezoelectrics (Uchino, 1997). Modern advances in organic materials have revealed that “soft” deformable dielectrics too hold tremendous potential to enable emerging technologies (Bar-Cohen, 2001; Carpi and Smela, 2009). At present, however, a major obstacle hindering the use of these soft active materials in actual devices is that they require — due to their inherent low permittivity — extremely high electric fields (>100 MV/m) to be actuated. Recent experiments have demonstrated that a promising solution to circumvent this limitation is to make composite materials, essentially by adding high-permittivity particles to the soft low-permittivity dielectrics (see, e.g., Zhang et al., 2002, 2007; Huang et al., 2005). Making composites out of hard deformable dielectrics has also proved increasingly beneficial for a broad range of applications (see, e.g., Akdogan et al., 2005). In this context, the objective of this work is to develop a microscopic field theory to describe, explain,

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and predict the macroscopic behavior of deformable dielectric composites directly in terms of their microscopic behavior. Motivated by the above-referenced experimental observations, the focus shall be on *finite electroelastic deformations* of composites with *two-phase particulate microstructures*.

To put the problem at hand in perspective, we recall that a complete macroscopic or phenomenological theory describing the quasistatic electromechanical behavior of elastic dielectrics has been available since the foundational paper of Toupin (1956) in the 1950s. Motivated by the renewed interest in electroactive materials of the last 15 years, this theory has been reformulated and presented in a variety of more convenient forms by a number of researchers including Dorfmann and Ogden (2005), McMeeking and Landis (2005), Vu and Steinmann (2007), Fosdick and Tang (2007), Xiao and Bhattacharya (2008), Suo et al. (2008). By contrast, microscopic or homogenization theories — needed to deal with composite materials — have not been pursued to nearly the same extent. Among the few results available, there are the well-established *linear* results for piezoelectric composites (see, e.g., Milton, 2002 and references therein) and the *nonlinear* result for electrostrictive composites of Tian et al. (2012), both within the restricted context of *small deformations* and *small electric fields*. Within the general context of *finite deformations* and *finite electric fields*, the only explicit results available in the literature appear to be those of deBotton et al. (2007) for the overall electrostrictive response of two-phase laminates; see also the finite element results of Li and Landis (2012). There is also the more recent work of Ponte Castañeda and Siboni (2012) wherein a decoupling approximation is proposed to model a special class of elastic dielectrics filled with mechanically rigid particles.

We begin this work in Section 2 by formulating the electroelastostatics problem defining the macroscopic response of two-phase elastic dielectric particulate composites under arbitrarily large deformations and electric fields. By means of an iterated homogenization procedure, we construct in Section 3 a solution for this problem for a specific but yet fairly general class of two-phase particulate (periodic or random) microstructures. This solution — given implicitly by the first-order nonlinear partial differential equation (35)–(36) and described in detail in Section 3.3 — constitutes the main result of this paper. It is valid for any choice of elastic dielectric behaviors for the matrix and particles, and any choice of one- and two-point correlation functions describing the underlying microstructure. For demonstration purposes, we spell out in Section 4 its specialization to the case when the matrix material is an ideal elastic dielectric. This result is further specialized in Section 4.1 to the case when the particles are ideal elastic dielectrics themselves, initially spherical in shape and distributed either isotropically (Section 4.1.1) or in chain-like formations (Section 4.1.2). In Section 5, we present sample results for the overall electrostrictive deformation that these particle-filled ideal dielectrics undergo when they are exposed to a uniaxial electric field. The aim there is to shed light on how the presence of filler particles — in terms of their elastic dielectric properties, volume fraction, and spatial distribution — affect the electrostrictive performance of deformable dielectrics. Finally, we provide in A, B, and C further details regarding the microscopic field theory developed in Section 3, including how it can be utilized to extract information on local fields; knowledge of local fields is of the essence, for instance, to probe the onset of electromechanical instabilities such as cavitation and electric breakdown.

2. Problem formulation

Microscopic description of the material. Consider a heterogeneous material comprising a continuous matrix filled by a statistically uniform (i.e., translation invariant) distribution of firmly bonded particles that occupies a domain Ω_0, with boundary $\partial\Omega_0$, in its undeformed stress-free configuration. The matrix is labeled as phase $r=1$, while the particles are collectively labeled as phase $r=2$. The domains occupied by each individual phase are denoted by $\Omega_0^{(1)}$ and $\Omega_0^{(2)}$, so that $\Omega_0=\Omega_0^{(1)}\cup\Omega_0^{(2)}$ and their respective volume fractions are given by $c_0^{(1)}\doteq|\Omega_0^{(1)}|/|\Omega_0|$ and $c_0^{(2)}\doteq|\Omega_0^{(2)}|/|\Omega_0|$. We assume that the characteristic size of the particles is much smaller than the size of Ω_0 and, for convenience, choose units of length so that Ω_0 has unit volume.

Material points are identified by their initial position vector $\mathbf{X}$ in Ω_0 relative to some fixed point. Upon the application of mechanical and electrical stimuli, the position vector $\mathbf{X}$ of a material point moves to a new position specified by $\mathbf{x}=\chi(\mathbf{X})$, where χ is a one-to-one mapping from Ω_0 to the deformed configuration Ω. We assume that χ is twice continuously differentiable, except possibly on the particles/matrix boundaries. The associated deformation gradient is denoted by $\mathbf{F}=\text{Grad}\,\chi$ and its determinant by $J=\det\mathbf{F}$.

Both the matrix ($r=1$) and the particles ($r=2$) are elastic dielectrics. We find it convenient to characterize their constitutive behaviors in a Lagrangian formulation by "total" free energies $W^{(r)}$ (suitably amended to include contributions from the Maxwell stress) per unit undeformed volume, as introduced by Dorfmann and Ogden (2005). In this work, such energy functions are assumed to be objective, differentiable, and, for definiteness, we shall use the deformation gradient $\mathbf{F}$ and Lagrangian electric field $\mathbf{E}$ as the independent variables[1]. It then follows that the first Piola–Kirchhoff stress tensor $\mathbf{S}$ and Lagrangian electric displacement $\mathbf{D}$ are given in terms of $\mathbf{F}$ and $\mathbf{E}$ simply by

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

[1] For our purposes here, it is equally convenient to use the Lagrangian electric displacement $\mathbf{D}$ as the electric independent variable instead of $\mathbf{E}$. For completeness, Appendix C includes a summary of results based on this alternative variable.