The Overall Elastic Dielectric Properties of Fiber-Strengthened/Weakened Elastomers

By employing recent results (Lopez-Pamies, O., 2014, “Elastic Dielectric Composite: Theory and Application to Particle-Filled Ideal Dielectrics,” J. Mech. Phys. Solids, 64, p. 6182 and Spinelli, S. A., Lefe`vre, V., and Lopez-Pamies, O., “Dielectric Elastomer Composites: A General Closed-Form Solution in the Small-Deformation Limit,” J. Mech. Phys. Solids, 83, pp. 263–284.) on the homogenization problem of dielectric elastomer composites, an approximate solution is generated for the overall elastic response of elastomers filled with a transversely isotropic distribution of aligned spheroidal particles in the classical limit of small deformations and moderate electric fields. The solution for such a type of dielectric elastomer composites is characterized by 13 (five elastic, two dielectric, and six electrostrictive) effective constants. Explicit formulae are worked out for these constants directly in terms of the elastic dielectric properties of the underlying elastomer and the filler particles, as well as the volume fraction, orientation, and aspect ratio of the particles. As a first application of the solution, with the objective of gaining insight into the effect that the addition of anisotropic fillers can have on the electromechanical properties of elastomers, sample results are presented for the case of elastomers filled with aligned cylindrical fibers. These results are confronted to a separate exact analytical solution for an assemblage of differential coated cylinders (DCC), wherein the fibers are polydisperse in size, and to full-field simulations of dielectric elastomer composites with cylindrical fibers of monodisperse size. These results serve to shed light on the recent experimental findings concerning the dielectric elastomers filled with mechanically stiff fibers. Moreover, they serve to reveal that high-permittivity liquid-like or vacuum fibers—two classes of filler materials yet to be explored experimentally—have the potential to significantly enhance the electrostriction capabilities of dielectric elastomers. [DOI: 10.1115/1.4031187]

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1 Introduction and Problem Setting

Through a certain asymptotic analysis within the context of the homogenization theory of Lopez-Pamies [1] in finite electroelasticstics, Spinelli et al. [2] have recently derived an exact closed-form solution for the overall electromechanical response of two-phase dielectric elastomer composites with specific yet fairly general types of (random or periodic) particulate microstructures1 in the classical limit of small deformations and moderate electric fields. The solution is given explicitly in terms of the electromechanical properties of the underlying matrix and fillers, and the one- and two-point correlation functions describing the microstructure. Opere citato, Spinelli et al. employed the solution to work out specific results for dielectric elastomers filled with isotropic distributions of spherical particles, a microstructure for which the required two-point correlation function takes on a particularly simple form. The purpose of this paper is to place on record corresponding analytical results for dielectric elastomers filled with transversely isotropic distributions of aligned spheroidal particles.

Microscopic description of dielectric elastomer composites. A dielectric elastomer composite is taken here to consist of a statistically uniform distribution of disconnected fillers that are perfectly bonded to a matrix. The domain occupied by the entire composite

in its ground state is denoted by Ω and its boundary by ∂Ω. Both phases, the matrix (r = 1) and the fillers (r = 2), are taken to be elastic dielectrics. Their constitutive behaviors are characterized by “total” free energies $W^{(r)}$ that are objective, isotropic functions of the deformation gradient $F$, and objective, isotropic, and even functions of the Lagrangian electric field $E$, i.e., $W^{(r)}(\mathbf{Q}, \mathbf{K}, \mathbf{E}) = W^{(r)}(\mathbf{F}, -\mathbf{E}) = W^{(r)}(\mathbf{F}, \mathbf{E})$ for all $\mathbf{Q}, \mathbf{K} \in Orth^r$. At a given material point $\mathbf{x}$, the first Piola-Kirchhoff stress $\mathbf{S}$ and the Lagrangian electric displacement $\mathbf{D}$ are then given in terms of $\mathbf{F}$ and $\mathbf{E}$ simply by Dorfman and Ogden [3]

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

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

where $\theta$ stands for the indicator function of the regions occupied by the fillers: $\theta(\mathbf{x}) = 1$ if $\mathbf{x}$ is inside a filler and 0 otherwise.

The macroscopic response. Granted the statistical uniformity of the distribution of the fillers and assuming that their characteristic size is much smaller than the length scale of Ω, the macroscopic length scale, the above-defined dielectric elastomer composite is expected to behave macroscopically as a homogeneous material. Its overall or macroscopic constitutive response is characterized by the relationship between the volume averages of the first Piola-Kirchhoff stress $\mathbf{S} = \int_{\Omega} \mathbf{S}(\mathbf{X}) d\mathbf{X}$ and electric displacement $\mathbf{D} = \int_{\Omega} \mathbf{D}(\mathbf{X}) d\mathbf{X}$ and the volume averages of the deformation gradient $\mathbf{F} = \int_{\Omega} \mathbf{F}(\mathbf{X}) d\mathbf{X}$ and electric field $\mathbf{E} = \int_{\Omega} \mathbf{E}(\mathbf{X}) d\Omega$ over Ω when the composite is subjected to affine boundary conditions [1].
relation can be conveniently written in terms of the total electroelastic free energy (per unit undeformed volume) of the composite

$$W(F, E) = \min_{E, F} \max_{\Omega} \frac{1}{V} \int_{\Omega} W(X, F, E) dX$$

as

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

where $\mathcal{K}$ and $\mathcal{E}$ in expression (3) denote sufficiently large sets of admissible deformation gradients $F$ and curl-free electric fields $E$ with prescribed volume averages $\bar{F}$ and $\bar{E}$. We note that the effective free energy function (3) is, by definition, an objective in $F$ and even in $E$ so that $W(F, E) = W(F, \bar{E}) = W(F, \bar{E}) = W(F, E)$ for all $\bar{Q} \in \text{Orth}$. Similar to the local free energy functions $W^{(i)}$ of the matrix and the fillers.

The classical limit of small deformations and moderate electric fields. The focus of this paper is on the classical asymptotic limit of small macroscopic deformations and moderate macroscopic electric fields. That is, letting $\zeta$ denote a vanishingly small parameter, the deformation measure $h = F - I$, with $I$ denoting the identity in the space of second-order tensors, is taken to be of $O(\zeta)$ while the electric field $E$ is of $O(\zeta^{1/2})$ (see, e.g., Refs. [4–6]). In this limit, the effective free energy function (3) takes the asymptotic form (in indicial notation)

$$W(F, E) = \frac{1}{2} \partial_{ijk} h_{ijkl} \bar{h}_{ijkl} - \frac{1}{2} E_{ij} \bar{\varepsilon}_{ij} + \frac{1}{2} \partial_{ijkl} \bar{M}_{ijkl} E_{ij} E_{kl} - E_{ij} \bar{E}_{ij} + O(\zeta^{3/2})$$

while the macroscopic constitutive relations (4) reduce to

$$\bar{S}_{ij} = \frac{\partial W}{\partial E}(F, E) = \bar{L}_{ijkl} \bar{h}_{ijkl} + \bar{M}_{ijkl} E_{ij} E_{kl} + O(\zeta^{3/2})$$

and

$$\bar{D}_{ij} = -\frac{\partial W}{\partial F}(F, E) = \bar{\varepsilon}_{ij} E_{ij} + O(\zeta^{3/2})$$

Here, $\bar{L}$ stands for the effective modulus of elasticity, $\bar{\varepsilon}$ denotes the effective permittivity, $\bar{M}$ is the effective electrostrictive tensor, and $\bar{r}$ represents the effective permittivity of second order. Note that $\bar{r}$ is absent from both asymptotic macroscopic constitutive relations (6) and (7). That is, in this classical limit of small macroscopic deformations and moderate macroscopic electric fields, the overall electromechanical response of dielectric elastomer composites is characterized by three effective tensors: the fourth-order tensor $\bar{L}$ describing their elasticity, the second-order tensor $\bar{\varepsilon}$ describing their permittivity, and the fourth-order tensor $\bar{M}$ describing their electrostrictive response. Because of the aforementioned objectivity of $W$ and its energy nature, these tensors are endowed with the following symmetries:

$$\bar{L}_{ijkl} = \bar{L}_{klij} = \bar{L}_{jikl} = \bar{L}_{ljjk}$$

$$\bar{\varepsilon}_{ij} = \bar{\varepsilon}_{ji}$$

$$\bar{M}_{ijkl} = \bar{M}_{klij} = \bar{M}_{lijk}$$

1.1 The Case of Transversely Isotropic Dielectric Elastomer Composites. The focus of this paper is furthermore on dielectric elastomer composites wherein the fillers are distributed with transverse isotropy in the undeformed configuration $\Omega$. Due to their microstructure, this type of composites exhibits an overall electromechanical behavior that is transversely isotropic. We shall denote their initial axis of symmetry by the unit vector $N$; as an illustrative example, Fig. 1 shows a schematic of a transversely isotropic dielectric elastomer composite with a distribution of axisymmetric fillers aligned in the direction of the axis of symmetry $N$. For this class of anisotropic dielectric elastomers, the effective free energy function (3) satisfies the material symmetry

![Fig. 1 Schematic of the microstructure of a transversely isotropic dielectric elastomer composite with initial axis of symmetry $N$](http://appliedmechanics.asmedigitalcollection.asme.org/)
when the fillers are spheroidal particles that are aligned and whose centers are distributed with transversely isotropic symmetry. To this end, we will employ the solution recently derived by Spinelli et al. [2] from the homogenization theory of Lopez-Pamies [1].

We begin in Sec. 2 by recalling the solution of Spinelli et al. [2] and spell out its specialization to transversely isotropic dielectric elastomers. In Sec. 3, we employ the latter to work out specific results for dielectric elastomers filled with transversely isotropic distributions of aligned spheroidal particles. Section 3 includes a discussion of the key features of the results and their specialization to limiting cases of practical interest. To shed light on recent experimental findings, we utilize in Sec. 4 the formulae worked out in Sec. 3 to examine the electrostriction response of the dielectric elastomers filled with cylindrical fibers that are mechanically stiff. With the aim of identifying what other type of fillers not yet utilized in experiments may potentially lead to enhanced behaviors, we also present corresponding sample results for dielectric elastomer composites with cylindrical fibers that are liquid-like and vacuous in mechanical behavior. Section 4 includes comparisons with a separate exact analytical solution and with full-field simulations, the details of which are presented in Appendices A and B.

2 The Solution of Spinelli et al.

For the physically relevant case when the matrix and the fillers are isotropic ideal elastic dielectrics, so that the free energy functions for the matrix and fillers are such that (see, e.g., Refs. [8,9])

\[
W^{(0)}(\mathbf{F}, \mathbf{E}) = \frac{1}{2} (F_{ij} - \delta_{ij}) L^{(i)}_{ijkl} (F_{kl} - \delta_{kl}) - \frac{1}{2} E_{ij} \epsilon^{(i)}_{ij} E_{ij} + (F_{ij} - \delta_{ij}) M^{(i)}_{ijkl} E_{kl} E_{ij} + \text{HOT} \tag{12}
\]

with

\[
\mathbf{L}^{(1)} = \frac{\partial^2 W^{(1)}}{\partial \mathbf{F}^2} \; (\mathbf{I}, 0) = 2\mu\mathbf{K} + (3\lambda + 2\mu)\mathbf{J}
\]

\[
\epsilon^{(1)} = -\frac{\partial W^{(1)}}{\partial \mathbf{E}^2} \; (\mathbf{I}, 0) = \mathbf{I}
\]

\[
M^{(1)} = \frac{\partial W^{(1)}}{\partial \mathbf{F}^2} \; (\mathbf{I}, 0) = \epsilon\mathbf{K} - \frac{\mathbf{e}}{2}\mathbf{J}
\]

\[
L^{(2)} = \frac{\partial^2 W^{(2)}}{\partial \mathbf{F}^2} \; (\mathbf{I}, 0) = 2\mu\mathbf{K} + (3\lambda + 2\mu)\mathbf{J}
\]

\[
\epsilon^{(2)} = -\frac{\partial W^{(2)}}{\partial \mathbf{E}^2} \; (\mathbf{I}, 0) = \epsilon\mathbf{I}
\]

\[
M^{(2)} = \frac{\partial W^{(2)}}{\partial \mathbf{F}^2} \; (\mathbf{I}, 0) = \epsilon\mathbf{K} - \frac{\epsilon}{2}\mathbf{J}
\]

where \(\lambda, \mu, \epsilon, \lambda_c, \mu_c, \epsilon_c\) stand, respectively, for the Lamé constants and the permittivities of the matrix and fillers, and the tensors \(\mathbf{K}, \mathbf{J}\) are given by

\[
K_{ijkl} = \frac{1}{2} \left[ \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl} \right]
\]

\[
J_{ijkl} = \frac{1}{3} \delta_{ij} \delta_{kl}
\]

the solution for the effective tensors \(\mathbf{L}, \bar{\epsilon}, \bar{M}\) generated in Ref. [2] reads as

\[
\bar{L}_{ijkl} = \frac{c}{2} \left( \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \delta_{ij} \delta_{kl} \right) + \frac{1}{2 c^2} \frac{\Delta \mu \Delta e}{\epsilon_0} \Delta \bar{L}_{ijkl} \bar{Q}_{kl} \bar{Q}_{ij}
\]

\[
\bar{\epsilon}_{ij} = \frac{1}{\epsilon_c} \left[ (1 - c) \mathbf{P}^{(e)} + \frac{\mathbf{K}}{2 \Delta \mu} + \frac{1}{3 \Delta \lambda + 2 \Delta \mu} \mathbf{J} \right]^{-1}
\]

\[
M_{ijkl} = \frac{c}{2} \left( \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \delta_{ij} \delta_{kl} \right) + \frac{1}{2 c^2} \frac{\Delta \mu \Delta e}{\epsilon_0} \Delta \bar{M}_{ijkl} \bar{Q}_{kl} \bar{Q}_{ij}
\]

Here, use has been made of the notation \(\Delta \mathbf{L} = \mathbf{L} - \mathbf{L}^{(1)}\), \(\Delta \mathbf{e} = \bar{\epsilon} - \epsilon^{(1)}\), \(\Delta \mathbf{L} = \mathbf{L}^{(2)} - \mathbf{L}^{(1)}\), \(\Delta \mathbf{e} = \epsilon^{(2)} - \epsilon^{(1)}\), \(\Delta \mathbf{M} = \mathbf{M}^{(2)} - \mathbf{M}^{(1)}\), \(\Delta \lambda = \lambda_c - \lambda\), \(\Delta \mu = \mu_c - \mu\), \(\Delta \epsilon = \epsilon_c - \epsilon\), \(c\) stands for the one-point correlation function, namely, the initial volume fraction of the fillers

\[
c = p^{(2)} = \frac{1}{|Q|} \int_{\Omega} \delta(X) \; dX
\]

and

\[
P^{(2)}_{ijkl} = \frac{1}{\mu} \left. \delta_{ik} \langle \xi_j \xi_k \rangle \right|_{(ij),(kl)} - \frac{\mu + \lambda}{\mu (2\mu + \lambda)} \langle \xi_i \xi_j \xi_k \xi_l \rangle
\]

where the bracketed subscripts denote symmetrization and the triangular brackets stand for the weighted average

\[
\langle \rangle = \int_{|\xi| = 1} \nu(\xi) d\xi
\]

For random microstructures, the weight function \(\nu(\xi)\) in Eq. (19) is given directly in terms of the two-point correlation function
\[
p^{(22)}(Y) = \frac{1}{|\Omega|} \int_{\Omega} \theta(Y + X) \theta(X) \, dX \tag{20}
\]

by
\[
\nu(\xi) = -\frac{1}{8\pi^2|\Omega|} \int_{\Omega} \left( \frac{p^{(22)}(X) - c^2}{1 - c^2} \delta''(\xi \cdot X) \right) \, dX \tag{21}
\]

where \(\delta''\) denotes the second derivative of the Dirac delta function with respect to its argument. For periodic microstructures, \(\nu(\xi)\) is given in terms of \(p^{(22)}\) by expression (13) in Ref. [2].

### 2.1 The Case of Transversely Isotropic Dielectric Elastomer Composites

The solution (15) is valid for any choice of the one- and two-point correlation functions \(p^{(2)} = \epsilon\) and \(p^{(22)}\) describing the microstructure. The latter enters expressions (15) through the microstructural tensors \(\langle \xi_i \xi_j \xi_k \xi_l \rangle\) and \(\langle \xi_i \xi_j \rangle\). For later use, it is expedient to recognize that for transversely isotropic microstructures these tensors admit the spectral form
\[
\langle \xi_i \xi_j \xi_k \xi_l \rangle = \frac{1}{4} \left( E_{ijkl}^{(1)} + E_{ijkl}^{(3)} + E_{ijkl}^{(2)} \right)
\]
\[
\langle \xi_i \xi_j \rangle = \frac{1}{2} \left( \delta_{ij} - N_i N_j \right) + \beta_2 N_i N_j
\]

where it is recalled that the tensors \(E^{(1)}\) through \(E^{(3)}\) are given in terms of the axis of symmetry \(N\) by relation (10), and that the triangular brackets stand for the weighted average (19) with the weight \(\nu(\xi)\) being given by expression (21) in terms of the two-point correlation function \(p^{(22)}\).

For transversely isotropic microstructures, in view of the relations (22) and (23), the solution (15) for the effective electromechanical tensors \(\mathbf{L}, \mathbf{e}, \mathbf{M}\) reduces indeed to the form (9) with the 13 effective electromechanical constants given by

\[
\begin{align*}
\bar{c}_L &= 2\lambda + 2\mu + \frac{2(\Delta\lambda + \Delta\mu)c}{\Delta\mu(3\lambda + 2\mu)} - \frac{2c(1 - c)[\Delta\lambda(\lambda + \mu) - \beta_2(\lambda + 2\mu)]}{\mu(\lambda + 2\mu)} \\
\bar{\sigma}_L &= 2\mu + \frac{2\mu\lambda(\lambda + 2\mu)c}{(\lambda + 2\mu)[\mu + (1 - c)\beta_1\Delta\mu] - (1 - c)\Delta\mu(\lambda + \mu)} \\
\bar{\epsilon}_L &= 2\mu + \frac{\epsilon c\Delta\epsilon}{\epsilon + (1 - c)\beta_1\Delta\epsilon}, \quad \bar{\epsilon}_i = \frac{\epsilon c\Delta\epsilon}{\epsilon + (1 - c)\beta_1\Delta\epsilon} \\
\bar{\epsilon}_M &= \frac{(1 - c)\Delta\epsilon^2(\Delta\lambda + \Delta\mu)}{2\epsilon c} + \frac{\Delta\epsilon^2[(\bar{c}_L - 2\lambda - 2\mu)\Delta\lambda - 2(\bar{e}_L - \lambda)(\Delta\lambda + \Delta\mu)]}{2\epsilon^2\Delta\mu\Delta\epsilon(3\lambda + 2\mu)} \\
&\quad + \left[\beta_1(\bar{c}_L - 2\lambda - 2\mu) + \beta_2(\bar{e}_L - \lambda)\right] \frac{1}{4\epsilon c} \left( \frac{1 + (1 + c)\Delta\mu}{\lambda + 2\mu} - \frac{\Delta\mu(\lambda + \mu)}{\mu(\lambda + 2\mu)} \right) \\
&\quad - \beta_2(1 - c)\Delta\epsilon^2(2\bar{e}_L - 2\lambda) \quad \frac{\Delta\epsilon^2[(\bar{c}_L - 2\lambda - 2\mu)\Delta\lambda - 2(\bar{e}_L - \lambda)(\Delta\lambda + \Delta\mu)]}{2\epsilon^2\Delta\mu\Delta\epsilon(3\lambda + 2\mu)} \\
&\quad + \frac{(1 - c)\Delta\epsilon^2(2\bar{e}_L - 2\lambda) + \beta_2(\bar{e}_L - \lambda)}{4\epsilon c} \left( \frac{1 + (1 + c)\Delta\mu}{\lambda + 2\mu} - \frac{\Delta\mu(\lambda + \mu)}{\mu(\lambda + 2\mu)} \right)
\end{align*}
\]

Here
\[
\begin{align*}
x_1 &= \frac{1}{2} \left( \mathbf{E}^{(1)}_{ijkl} \xi_i \xi_j \xi_k \xi_l \right) \\
x_2 &= \frac{1}{2} \left( \mathbf{E}^{(2)}_{ijkl} \xi_i \xi_j \xi_k \xi_l \right) \\
x_3 &= \frac{1}{2} \left( \mathbf{E}^{(3)}_{ijkl} \xi_i \xi_j \xi_k \xi_l \right)
\end{align*}
\]

\begin{align*}
\beta_1 &= \frac{1}{2} \left( 1 - \xi_i N_i N_j \right), \quad \beta_2 = \left( \xi_i N_i N_j \right)
\end{align*}

where it is recalled that the tensors \(E^{(1)}\) through \(E^{(3)}\) are given in terms of the axis of symmetry \(N\) by relation (10), and that the triangular brackets stand for the weighted average (19) with the weight \(\nu(\xi)\) being given by expression (21) in terms of the two-point correlation function \(p^{(22)}\).
\[ \bar{\varepsilon}_M = \varepsilon + \frac{x_1(1 - c)\Delta e^2}{4\varepsilon} + \frac{\Delta^2(\bar{\varepsilon}_L - 2\mu)}{2c^2\Delta\mu\Delta e} \left\{ 1 + x_1(1 - c)\Delta e \right\} \left\{ 1 + \frac{(1 + c)\Delta e}{\lambda + 2\mu} \right\} \]

\[ \bar{\varepsilon}_M = \varepsilon + \frac{x_1(1 - c)\Delta e^2}{2\varepsilon} + \frac{\Delta^2(\bar{\varepsilon}_L - 2\mu)}{2c^2\Delta\mu\Delta e} \left\{ 1 + \frac{\beta_1(1 - c)\Delta e(\Delta e - 2\mu)}{2\mu\Delta e} \right\} \times \left\{ 1 + \frac{(1 + c)\Delta e}{\lambda + 2\mu} \right\} \]

\[ \bar{\varepsilon}_M = \left( 1 - c \right)\Delta e^2 \left( \bar{\varepsilon}_L - \lambda - 2\mu \right) + \frac{\beta_1(1 - c)\Delta e^2(\Delta e - 2\mu)}{2c^2\Delta\mu\Delta e^2} \left\{ \bar{\varepsilon}_L - 2\lambda - 2\mu \right\} \left\{ 1 + \frac{(1 + c)\Delta e}{\lambda + 2\mu} \right\} \]

\[ \bar{\varepsilon}_M = \left( 1 - c \right)\Delta e^2 \left( \bar{\varepsilon}_L - \lambda - 2\mu \right) + \frac{\beta_1(1 - c)\Delta e^2(\Delta e - 2\mu)}{2c^2\Delta\mu\Delta e^2} \left\{ \bar{\varepsilon}_L - 2\lambda - 2\mu \right\} \left\{ 1 + \frac{(1 + c)\Delta e}{\lambda + 2\mu} \right\} \]

where the notation \( \Delta e = \bar{\varepsilon} - \varepsilon \), \( \bar{\varepsilon}_L = \bar{\varepsilon} - \varepsilon \).

\[ \Upsilon = \left( \frac{\Delta e + \Delta \mu}{\Delta \mu(3\lambda + 2\mu)} + \frac{(1 - c)\xi_2(\lambda + \mu) - \xi_2(\lambda + 2\mu)}{\mu(\lambda + 2\mu)} \right) \left( \frac{\Delta e + 2\mu}{\Delta \mu(3\lambda + 2\mu)} - \frac{(1 - c)\xi_2(\lambda + \mu) - \xi_2(\lambda + 2\mu)}{\mu(\lambda + 2\mu)} \right) \]

\[ \nu(\xi) = \frac{\det Z}{4\pi|Z|^2} \text{ with } Z = I + (\omega - 1)N \otimes N \]

3 Approximations (based on classical linear estimates) for the overall electromechanical response of dielectric elastomer composites with this type of microstructures has been proposed earlier by Li and Rao [17] and by Siboni and Ponte Castañeda [18], the latter being specific for particles that are mechanically rigid.

3 Application to Dielectric Elastomers Filled With Aligned Spherical Particles

The majority of experimental studies on dielectric elastomer composites to date have been concerned with fillers that are roughly spherical in shape (see, e.g., Refs. [11–13]). There are, however, a number of studies (see, e.g., Refs. [14–16]) that have indicated that anisotropic fillers in the form of short (needle-like) and long (cylindrical) fibers can potentially lead to even larger enhancements than those endowed by spherical fillers. In this section, motivated in part to shed light on these findings, we work out the specialization of the effective electromechanical constants (24) to the case of dielectric elastomer composites wherein the underlying fillers are spherical in shape and are all aligned in the same direction. In order to favor analytical tractability, we consider that the centers of the particles are distributed with “spherical” symmetry and that the “aspect ratio” of this spherical distribution is the same as the aspect ratio of the particles\(^3\) [10]. For this class of microstructures, the weight function (21) reduces rather simply to [1,10].
and a transversely isotropic distribution of aligned cylindrical fibers with circular cross section (corresponding to \( \omega = \infty \)).

Now, given the weight function (26), it is straightforward to determine the corresponding parameters (23) needed in the evaluation of the effective constants (24). They read as follows:

\[
x_1 = \frac{\omega^2(2\omega^2 + 1) - \rho(\omega)(4\omega^2 - 1)}{4(\omega^2 - 1)^2}, \quad x_2 = \frac{2 + \omega^2 - 3\rho(\omega)}{2(\omega^2 - 1)^2}
\]

\[
x_3 = \frac{\rho(\omega)(2\omega^2 + 1) - 3\omega^2}{2(\omega^2 - 1)^2}
\]

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

(27)

where

\[
\rho(\omega) = \frac{\omega \sinh^{-1}(\sqrt{\omega^2 - 1})}{\sqrt{\omega^2 - 1}} = \frac{\omega \sin^{-1}(\sqrt{1 - \omega^2})}{\sqrt{1 - \omega^2}}
\]

(28)

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

Substitution of relations (27) in expressions (24) renders the main result of this paper, namely, the 13 effective constants associated with the transversely isotropic distribution of aligned spheroidal particles (24) in the three effective tensors (9) that characterize the overall electromechanical response of dielectric elastomers filled with a transversely isotropic distribution of aligned spheroidal particles:

\[
\begin{align*}
\bar{\sigma}_L &= 2\lambda + 2\mu + \frac{2(\Delta \lambda + \Delta \mu)\varepsilon}{\Delta \mu(3\Delta \lambda + 2\Delta \mu)} \left[ 2(1 - c) \left[ 2(\lambda + \mu) - 3(3\lambda + 5\mu) \omega^2 + (2\omega^2(\lambda + \mu) - \mu) \rho \right] \right] \\
\bar{\sigma}_M &= \frac{\Delta \varepsilon}{2} \left[ \frac{\Delta \varepsilon}{2(\omega^2 - 1)} + \frac{c \epsilon \Delta \sigma(\varepsilon^2)}{\varepsilon^2} \right] \\
\bar{\sigma}_M &= \frac{\Delta \varepsilon}{2} \left[ \frac{\Delta \varepsilon}{2(\omega^2 - 1)} + \frac{c \epsilon \Delta \sigma(\varepsilon^2)}{\varepsilon^2} \right]
\end{align*}
\]

Fig. 2 Schematic of the microstructure of a dielectric elastomer filled with aligned spheroidal particles of aspect ratio \( \omega \) distributed with spheroidal symmetry of the same aspect ratio \( \omega \). The lower half of the figure shows the limiting cases of (a) an isotropic distribution of spherical particles (corresponding to \( \omega = 1 \)) and (b) a transversely isotropic distribution of aligned cylindrical fibers with circular cross section (corresponding to \( \omega = \infty \)).
\[
\bar{\varepsilon}_M = \varepsilon + \frac{(1-c)\Delta \varepsilon^2}{16c\varepsilon(\omega^2-1)^2}\left[\frac{1}{4} + \frac{(\bar{\varepsilon}_L - 2\mu)}{2c\Delta \mu}\left(1 + \frac{(1+c)\Delta \mu}{\lambda + 2\mu}\right) \right]
\]
\[
+ \frac{\Delta \varepsilon^2(\bar{\varepsilon}_L - 2\mu)}{2c^2\Delta \mu \Delta \varepsilon}\left[1 + \frac{(1-c)\Delta \mu(\lambda + 3\mu)(\Delta \varepsilon + c\Delta \varepsilon)}{4\mu\Delta \varepsilon(\lambda + 2\mu)}\right] \frac{1}{1 + \frac{\lambda^2(2 + \omega^2 - 3\rho) + \mu(6 + (4\rho - 3)\omega^2 - 7\rho)}{2(\omega^2 - 1)^2(\lambda + 3\mu)}},
\]
\[
\bar{\varepsilon}_M = \varepsilon + \frac{\Delta \varepsilon^2}{2c^2\Delta \mu \Delta \varepsilon}\left[\frac{1}{4} + \frac{(1-c)\Delta \mu}{\Delta \varepsilon}\left(\frac{\rho(2\omega^2 + 1) - 3\omega^2}{\rho(2\omega^2 + 1) - 3\omega^2}\right) \right]
\]
\[
- \frac{(1-c)\Delta \varepsilon(\bar{\varepsilon}_L - 2\mu)}{2c^2\Delta \mu(3\lambda + 2\Delta \varepsilon)}\left[\bar{\varepsilon}_L - 2\lambda + 2(\bar{\varepsilon}_L - \lambda)\left(1 + \frac{\rho(2\omega^2 + 1) - 3\omega^2}{\rho(2\omega^2 + 1) - 3\omega^2}\right)\right] - \frac{\Delta \lambda}{\Delta \mu}\left(\frac{1}{\mu\Delta \varepsilon}(\lambda + 2\mu)\right),
\]
\[
\bar{\varepsilon}_M = \varepsilon + \frac{\Delta \varepsilon^2}{2c^2\Delta \mu(3\lambda + 2\Delta \varepsilon)}\left[\bar{\varepsilon}_L - 2\lambda - 2\mu + (\bar{\varepsilon}_L - \lambda)\left(1 - \frac{\rho(2\omega^2 + 1) - 3\omega^2}{\rho(2\omega^2 + 1) - 3\omega^2}\right)\right]
\]
\[
\times \frac{\lambda + 2\mu + (1+c)\Delta \mu}{2c\Delta \mu} - \frac{(1+c)\Delta \varepsilon(\bar{\varepsilon}_L - \lambda)}{\mu\Delta \varepsilon}\left(1 + \frac{2\mu(3\lambda + 2\Delta \varepsilon)}{\mu(\lambda + 2\mu)}\right)
\]
\[
(29)
\]

where now
\[
Y = \left(\frac{\Delta \lambda + \Delta \mu}{\Delta \mu(3\lambda + 2\Delta \varepsilon)}\right) + \frac{(1-c)\rho(2\omega^2 + \lambda + 2\mu) - (3\lambda + 5\mu)\omega^2 + 2\mu}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2}
\]
\[
- \frac{\Delta \lambda + \Delta \mu}{\Delta \mu(3\lambda + 2\Delta \varepsilon)} + \frac{(1-c)\rho(2\omega^2 + 3\lambda + 5\mu)\omega^2 + 2\mu(2\omega^2 + 3\lambda + 5\mu)}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2}
\]
\[
- \frac{\Delta \lambda}{\Delta \mu(3\lambda + 2\Delta \varepsilon)} - \frac{(1-c)\rho(2\omega^2 + 1) - 3\omega^2}{2\mu(\lambda + 2\mu)(\omega^2 - 1)^2},
\]
\[
(30)
\]

and it is recalled that \(\Delta \lambda = \lambda_0 - \lambda, \Delta \mu = \mu_0 - \mu, \Delta \varepsilon = \varepsilon_0 - \varepsilon, \Delta \varepsilon = \bar{\varepsilon}_0 - \bar{\varepsilon}, \) and \(\Delta \varepsilon_0 = \bar{\varepsilon}_0 - \bar{\varepsilon}.
\]

### 3.1 Some Limiting Cases of Practical Interest

- **Rigid particles and incompressible elastomers.** In the limit of rigid particles when \(\mu = \lambda = \infty\) and incompressible elastomers when \(\omega = \infty\), the effective electromechanical constants (29) reduce to

\[
\bar{\varepsilon}_L = \bar{\varepsilon}_L = \infty, \quad \bar{\varepsilon}_L = 2\mu \varepsilon_0^2 - 1)^2 (1-c)(3\rho + 2\omega^2 - 5\omega^2), \quad \bar{\varepsilon}_L = 2\mu + \frac{4c\mu(\omega^2 - 1)^2}{(1-c)(\omega^2 + 2\omega^2 - 3\omega^2)}, \quad \bar{\varepsilon}_L = \infty
\]
\[
\bar{\varepsilon}_L = \bar{\varepsilon}_L = \infty, \quad \bar{\varepsilon}_L = 2\mu \varepsilon_0^2 - 1)^2 (1-c)(3\rho + 2\omega^2 - 5\omega^2), \quad \bar{\varepsilon}_L = 2\mu + \frac{4c\mu(\omega^2 - 1)^2}{(1-c)(\omega^2 + 2\omega^2 - 3\omega^2)}, \quad \bar{\varepsilon}_L = \infty
\]
\[
\bar{\varepsilon}_L = \bar{\varepsilon}_L = \infty, \quad \bar{\varepsilon}_L = 2\mu \varepsilon_0^2 - 1)^2 (1-c)(3\rho + 2\omega^2 - 5\omega^2), \quad \bar{\varepsilon}_L = 2\mu + \frac{4c\mu(\omega^2 - 1)^2}{(1-c)(\omega^2 + 2\omega^2 - 3\omega^2)}, \quad \bar{\varepsilon}_L = \infty
\]

\[
\bar{\varepsilon}_L = \bar{\varepsilon}_L = \infty, \quad \bar{\varepsilon}_L = 2\mu \varepsilon_0^2 - 1)^2 (1-c)(3\rho + 2\omega^2 - 5\omega^2), \quad \bar{\varepsilon}_L = 2\mu + \frac{4c\mu(\omega^2 - 1)^2}{(1-c)(\omega^2 + 2\omega^2 - 3\omega^2)}, \quad \bar{\varepsilon}_L = \infty
\]
These results are relevant for standard dielectric elastomers, which are typically nearly incompressible, filled with ceramic or metallic particles.

- **Liquid-like particles and incompressible elastomers.** The limiting values $\mu_0 = 0$, $\lambda = \infty$, and $\lambda = \infty$ correspond to particles that are liquid-like (incompressible with vanishingly small shear resistance) and elastomers that are incompressible. Granted these values, the effective electromechanical constants (29) reduce to

\[
\ddot{\bar{\varepsilon}}_M = \frac{\varepsilon}{2} + \frac{\Delta \bar{\varepsilon}_i^2 [\omega^2 (2\rho - 4c\rho + c - 3) + (1 + c)\rho + 2c\omega^2]}{8\epsilon_0 (\omega^2 - 1)^2} \\
\ddot{\bar{\kappa}}_M = \frac{\varepsilon}{2} + \frac{\Delta \bar{\varepsilon}_i^2 [\omega^2 (2\rho + 2c\rho - c - 3) + (1 - 5c)\rho + 4c]}{8\epsilon_0 (\omega^2 - 1)^2}
\]

(31)

The results (32) are relevant for standard dielectric elastomers filled with common fluids, such as water, or eutectic alloys, such as Galinstan.

- **Porous dielectric elastomers.** Another limit of practical relevance contained in the result (29) is that of porous dielectric elastomers. This corresponds to setting $\mu_0 = \lambda = 0$ and $\epsilon = \epsilon_0$, with $\epsilon_0 = 8.85 \times 10^{-12}$ F/m denoting the permittivity of vacuum. Assuming that the underlying elastomer is incompressible, so that $\lambda = \infty$, the effective electromechanical constants (29) in this context reduce to

\[
\ddot{\bar{\varepsilon}}_L = \ddot{\bar{\kappa}}_L = \infty, \quad \ddot{\bar{\varepsilon}}_L = 2\mu - \frac{8\epsilon_0 (\omega^2 - 1)^2}{2(1 + c)\omega^4 - (3 + 5c)\omega^2 - 3(1 - c)\rho + 4} \\
\ddot{\bar{\kappa}}_L = 2\mu - \frac{4\epsilon_0 (\omega^2 - 1)^2}{3(1 - c)\rho (\omega^2 + 1) + (1 + c)\omega^4 - (7 - 3c)\omega^2 + 2c} \\
\ddot{\bar{\varepsilon}}_t = \frac{\Delta \bar{\varepsilon}_i^2 (\rho - 1)}{2\epsilon_0 (\omega^2 - 1)^2} \left[ \frac{1 + c}{(2\rho - 1)} + \frac{\omega^2 [2c(\rho - 3) - (2 - 2c)]}{2c\epsilon_0 (\omega^2 - 1)^2} \right] \\
\ddot{\bar{\kappa}}_M = \frac{\varepsilon}{2} + \frac{\Delta \bar{\varepsilon}_i^2 [1 + \omega^2 c(2\rho - 3) - (2 - 2c)]}{8(1 - c)\epsilon_0 (\rho - 1) + (\omega^2 - 1)^2} \\
\ddot{\bar{\varepsilon}}_M = \frac{\varepsilon}{2} + \frac{\Delta \bar{\varepsilon}_i^2}{2\epsilon_0 (\omega^2 - 1)^2} \left[ \frac{1 + c}{(2\rho - 1)} + \frac{\omega^2 [2c(\rho - 3) - (2 - 2c)]}{2c\epsilon_0 (\omega^2 - 1)^2} \right] \\
\ddot{\bar{\kappa}}_M = \frac{\varepsilon}{2} + \frac{\Delta \bar{\varepsilon}_i^2}{4\epsilon_0 (\omega^2 - 1)^2} \left[ \frac{1 + c}{(2\rho - 1)} + \frac{\omega^2 [2c(\rho - 3) - (2 - 2c)]}{2c\epsilon_0 (\omega^2 - 1)^2} \right]
\]

(32)
\[ \bar{c}_L = \frac{2(1-c)\mu_c}{\epsilon_c} \left[ 1 + \frac{(2\rho - 1)(3\rho - \omega^2 - 2) - 6c(\rho - 1)^2}{3(1-c)(2\omega^2 + 1)\rho - (5 - 9c)\omega^2 - 2\omega^4 - 2} \right] \]

\[ \bar{d}_L = \frac{(1-c)\mu_c}{\epsilon_c} \left[ 6(1-c)\rho^2 + 4\rho \left( 1 + 3c \right) \omega^2 - 1 \right] - 2(1 + 3c)\omega^4 - 4\omega^2 \]

\[ \bar{c}_L = \frac{2(1-c)\mu_c}{\epsilon_c} \left[ (1-c)\rho^2 + \rho \left( 2 - 3c \right) \omega^2 - 2 - 2(1 - 3c)\omega^2 - \omega^4 \right] \]

\[ \bar{\eta}_L = 2\mu + \frac{8c\mu(\omega^2 - 1)^2}{3(1-c)\rho - 2(1+c)\omega^2 + (3+5c)\omega^2 - 4} \]

\[ \bar{f}_L = 2\mu - \frac{4c\mu(\omega^2 - 1)^2}{3(1-c)(\omega^2 + 1)} + (1 + c)\omega^4 - (7 - 3c)\omega^2 + 2c \]

\[ \bar{\eta}_L = \frac{2(1-c)\mu_c}{\epsilon_c} \left[ (1-c)\rho^2 + \rho \left( 2 - 3c \right) \omega^2 - 2 - 2(1 - 3c)\omega^2 - \omega^4 \right] \]

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

\[ \bar{d}_M = \frac{e}{2} (1 - 2c)\Delta \bar{e} \left\{ 1 + \frac{3(1-c)\Delta \rho (\rho - 1)}{2(1-2c)(\omega^2 - 1)^2} \right\} \]

\[ \bar{c}_M = \frac{e + \frac{6\Delta \bar{e}^2(\omega^2 - 1)^2}{4c^2\Delta \bar{e}(\omega^2 - 1)^2}}{1 - \frac{3(1-c)\Delta \rho (\rho - 1)}{2(1-2c)(\omega^2 - 1)^2}} \]

The sets of expressions (31), (32), (33) correspond to dielectric elastomer composites with particles and elastomers that exhibit limiting constitutive behaviors. The result (29) also contains special cases associated with limiting geometries of the particles and their spatial distribution. Below, we spell out two of them.
Isotropic distribution of spherical particles. When the aspect ratio $\omega = 1$, the underlying microstructure reduces to an isotropic distribution of spherical particles (see Fig. 2(a)), and the 13 effective constants (29) reduce to just five (two elastic, one dielectric, and two electrostrictive) independent effective constants. This problem was addressed in Sec. 5 of Ref. [2] and the interested reader is referred to that reference for a detailed discussion. For completeness, we record here the specialization of the effective electromagnetic constants (29)

\[ \bar{c}_L = 2 \bar{\lambda} + 2 \bar{\mu}, \quad \bar{d}_L = \bar{\lambda} + 2 \bar{\mu} \]
\[ \bar{e}_L = 2 \bar{\mu}, \quad \bar{g}_L = \bar{\lambda} \]
\[ \bar{c}_M = \frac{1}{3} \bar{m}_K + \frac{2}{3} \bar{m}_J, \quad \bar{d}_M = \frac{2}{3} \bar{m}_K + \frac{1}{3} \bar{m}_J \]
\[ \bar{e}_M = \bar{m}_K, \quad \bar{g}_M = \bar{m}_J = \frac{1}{3} (\bar{m}_J - \bar{m}_K) \]

where

\[ \bar{\mu} = \mu + \frac{15 c \mu (\lambda + 2 \mu) \Delta \mu}{15 \mu (\lambda + 2 \mu) + 2(1 + c)(3\lambda + 8\mu) \Delta \mu} \]
\[ \bar{\lambda} = \lambda + c \Delta \lambda - \frac{c (1 - c) (3\Delta \lambda + 2 \Delta \mu)^2}{9 (\lambda + 2 \mu) + 3 (1 - c)(3\lambda + 2 \Delta \mu)} \]
\[ + \frac{4 c (1 - c) (3\lambda + 8\mu) \Delta \mu^2}{45 \mu (\lambda + 2 \mu) + 6 (1 - c)(3\lambda + 8\mu) \Delta \mu} \]
\[ \bar{\varepsilon} = \varepsilon + \frac{3 c \varepsilon \Delta \varepsilon}{5 \varepsilon + (1 - c) \Delta \varepsilon} \]
\[ \bar{m}_K = \varepsilon + \frac{3 c \varepsilon \Delta \varepsilon [15 \varepsilon + 2 (1 - c) \Delta \varepsilon]}{5 [3 \varepsilon + (1 - c) \Delta \varepsilon]^2} \]
\[ + \frac{9 c (1 - c) \varepsilon \Delta \varepsilon \Delta \mu^2}{5 [3 \varepsilon + (1 - c) \Delta \varepsilon]^2} \]
\[ \times \frac{(3 - 1 \varepsilon) \lambda + (13 - 3 \varepsilon) \mu}{15 \mu (\lambda + 2 \mu) + 2 (1 - c)(3\lambda + 8\mu) \Delta \mu} \]
\[ \bar{m}_J = -\frac{\varepsilon}{2} + \frac{3 c \varepsilon \Delta \varepsilon}{2 [3 \varepsilon + (1 - c) \Delta \varepsilon]} + \frac{c (1 - c) \varepsilon \Delta \varepsilon}{[3 \varepsilon + (1 - c) \Delta \varepsilon]^2} \]
\[ \times \frac{9(3\lambda + 2 \Delta \mu)(3 \varepsilon + \Delta \varepsilon)}{6 (\lambda + 2 \mu) + 2 (1 - c)(3\lambda + 2 \Delta \mu)} \]

Transversely isotropic distribution of cylindrical fibers with circular cross section. When the aspect ratio $\omega = \infty$, the underlying microstructure reduces to a transversely isotropic distribution of cylindrical fibers with circular cross section (see Fig. 2(b)). In this limiting case, expressions (29) simplify to

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

A common approach to characterize the electromechanical properties of dielectric elastomers (filled or unfilled) is to measure their deformation while they are subjected to a uniaxial electric field in the absence of stress (see, e.g., Sec. 2.25 in Refs. [4,19]). Such an electrically induced deformation is usually referred to as **electrostriction**.

From the constitutive relation (6), it follows that, in the absence of stress where $\mathbf{S} = 0$, the macroscopic electrostriction $\Pi$ induced in a dielectric elastomer composite by a uniform macroscopic electric field $\mathbf{E}$ is formally given by

$$\Pi = -\mathbf{L}^{-1} \tilde{\mathbf{M}} \mathbf{E} \otimes \mathbf{E}$$  \hspace{1cm} (37)

Now, exploiting the fact that the effective modulus of elasticity here is of the transversely isotropic form (9), its inverse $\mathbf{L}^{-1}$ can be conveniently written as [7]

$$\mathbf{L}^{-1} = \frac{d_{L}}{c_{L} d_{L} - 2g_{L}} \mathbf{E}^{(1)} + \frac{c_{L}}{c_{L} d_{L} - 2g_{L}} \mathbf{E}^{(2)} + \frac{1}{\tilde{e}_{L}} \mathbf{E}^{(3)} + \frac{1}{\tilde{e}_{L}} \mathbf{E}^{(4)} - \frac{\tilde{a}_{L}}{c_{L} d_{L} - 2g_{L}} \mathbf{E}^{(5)} + \mathbf{E}^{(6)}$$  \hspace{1cm} (38)

where it is recalled that the tensors $\mathbf{E}^{(1)}$ through $\mathbf{E}^{(6)}$ are defined by relations (10). Upon direct use of this expression, the expression (9) for the effective electrostrictive tensor $\tilde{\mathbf{M}}$, and some algebraic manipulation, the electrostriction (37) can be written more explicitly as

$$\Pi = -\left( \frac{d_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(1)} + \left( \frac{c_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(2)} + \frac{\tilde{a}_{L}}{\tilde{e}_{L}} \mathbf{E}^{(3)} + \frac{\tilde{g}_{L}}{\tilde{e}_{L}} \mathbf{E}^{(4)} + \left( \frac{c_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(5)} + \mathbf{E}^{(6)}$$  \hspace{1cm} (39)

It is of note that this relation depends on all five effective elastic constants ($c_{L}, d_{L}, \tilde{a}_{L}, \tilde{e}_{L}, \tilde{g}_{L}$) and all six effective electrostrictive constants ($\tilde{c}_{M}, \tilde{d}_{M}, \tilde{e}_{M}, \tilde{f}_{M}, \tilde{g}_{M}, \tilde{h}_{M}$). It is also worth remarking that when the applied electric field $\mathbf{E}$ is aligned with the axis of symmetry $\mathbf{N}$ ($\mathbf{E} = \mathbf{N}$) or orthogonal to it ($\mathbf{N} \cdot \mathbf{E} = 0$), the electrostriction (39) reduces to

$$\Pi = -\left( \frac{d_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(1)} + \left( \frac{c_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(2)} + \frac{\tilde{a}_{L}}{\tilde{e}_{L}} \mathbf{E}^{(3)} + \frac{\tilde{g}_{L}}{\tilde{e}_{L}} \mathbf{E}^{(4)} + \left( \frac{c_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(5)} + \mathbf{E}^{(6)}$$  \hspace{1cm} (40)

for the former and to

$$\Pi = \frac{1}{2} \left( \frac{d_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(1)} \mathbf{E}^{(2)} + \frac{\tilde{a}_{L}}{\tilde{e}_{L}} \mathbf{E}^{(3)} + \frac{\tilde{g}_{L}}{\tilde{e}_{L}} \mathbf{E}^{(4)} + \left( \frac{c_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \right) \mathbf{E}^{(5)} \mathbf{E}^{(6)}$$  \hspace{1cm} (41)

for the latter. As opposed to the general result (39), the “aligned” electrostriction (40) depends only on three effective elastic constants ($c_{L}, d_{L}, \tilde{g}_{L}$) and two effective electrostrictive constants ($\tilde{c}_{M}, \tilde{h}_{M}$). Similarly, the “orthogonal” electrostriction (41) depends only on four effective elastic constants ($c_{L}, d_{L}, \tilde{a}_{L}, \tilde{g}_{L}$) and three effective electrostrictive constants ($\tilde{c}_{M}, \tilde{e}_{M}, \tilde{g}_{M}$).

### 4 Sample Results

In addition to their theoretical value in providing a rigorous analytical solution for a fundamental nonlinear coupled problem, the 13 effective electromechanical constants (29) provide a formidable tool to gain insight into how the addition of anisotropic particles may enhance the electromechanical properties of dielectric elastomers. In this section, for illustration purposes, we seek to gain some of that insight via sample results.

We restrict ourselves to examining dielectric elastomer composites wherein the fillers are aligned cylindrical fibers with circular cross section; in this case, $\omega = \infty$ and the general formulae (29) reduce to expressions (36). The practical motivation to consider such a type of composites stems from the fact that they can be readily fabricated by means of modern synthesis/manufacturing processes (see, e.g., Refs. [16,20,21]). With the objective of presenting results that are directly relatable to standard experimental measurements, we further restrict our attention to examining the electrostriction of these composites when subjected to an electric field that is orthogonal to the fibers (this involves only a subset of the thirteen effective electromechanical constants that characterize their electromechanical behavior). As shown schematically by Fig. 3, we choose the $e_{3}$ axis of the laboratory frame of reference to coincide with the direction of the fibers $\mathbf{N}$, while the $e_{1}$ axis is chosen to coincide with the direction of the applied electric field $\mathbf{E} = E e_{1}$. Given this choice of frame of reference, the electrostriction $\Pi$ in the composite takes the simple diagonal form (cf. Eq. (41))

$$\Pi = \Pi_{11} e_{1} \otimes e_{1} + \Pi_{22} e_{2} \otimes e_{2} + \Pi_{33} e_{3} \otimes e_{3}$$  \hspace{1cm} (42)

with

$$\Pi_{11} = \frac{d_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \mathbf{E}^{2}$$  \hspace{1cm} (43)

$$\Pi_{22} = \frac{c_{L} \tilde{a}_{L} - 2g_{L} \tilde{g}_{L}}{c_{L} d_{L} - 2g_{L}} \mathbf{E}^{2}$$  \hspace{1cm} (44)

$$\Pi_{33} = \frac{\tilde{a}_{L} \tilde{c}_{M} - 2g_{L} \tilde{g}_{M}}{c_{L} d_{L} - 2g_{L}} \mathbf{E}^{2}$$  \hspace{1cm} (45)

where it is recalled that, for the case of cylindrical fibers of interest here, the effective electromechanical constants $c_{L}, d_{L}, \tilde{a}_{L}, \tilde{g}_{L}, \tilde{c}_{M}, \tilde{e}_{M}, \tilde{g}_{M}$ are given explicitly by expressions (36).
In the next three subsections, we present and discuss numerical results for the electrostriction components (43) for dielectric elastomer composites wherein the elastomer, has permittivity \( \varepsilon = 3.2\varepsilon_0 \) and Lamé constant \( \lambda = \infty \), while the fibers exhibit various permittivities and mechanical behaviors ranging from stiff, to liquid-like, to vacuous. To better illustrate the effect that the addition of fibers has on the electrostriction performance of elastomers, the results are presented in terms of the ratios \( \Pi_{11}/H_{11}^m, \Pi_{22}/H_{22}^m, \Pi_{33}/H_{33}^m \), where

\[
H_{11}^m = H_{33}^m = \frac{\varepsilon}{6\mu} \varepsilon^2 \quad \text{and} \quad H_{22}^m = -\frac{\varepsilon}{3\mu} \varepsilon^2 \quad \text{(44)}
\]

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

To aid the discussion in all of the results that follow, we include comparisons with a separate exact analytical solution, as well as with full-field solutions constructed by means of the finite-element (FE) method. The analytical solution corresponds to a dielectric elastomer filled with a special type of transversely isotropic distribution of cylindrical fibers—the so-called differential dielectric elastomer filled with a special type of transversely isotropic distribution of rigid fibers with circular cross section(s); see Fig. 3. Results are shown for fibers with permittivities \( \varepsilon_l = 4\varepsilon_0 \) and \( 10^4\varepsilon_0 \), as functions of the volume fraction of fibers \( c \). The solid line corresponds to the theoretical result (45). The dashed line corresponds to the response of a DCC assemblage of polydisperse fibers (see Appendix A), while the solid circles correspond to FE simulations of a dielectric elastomer composite with monodisperse fibers (see Appendix B).

4.1 Stiff Fibers. We begin by examining dielectric elastomer composites with fibers that are mechanically stiff. For definiteness, given that most materials commonly utilized as fibers (e.g., carbon) are much stiffer than elastomers, we take the fibers to be rigid, \( \mu_c = \lambda_c = \infty \). Due to the overall rigidity in the direction of the fibers and the overall incompressibility of the resulting composite, the electrostriction components (43), normalized by the matrix electrostriction components (44), take the particular simple form

\[
\frac{\Pi_{11}}{H_{11}^m} = \frac{3(1-c)}{4(1+c)} \frac{[8\varepsilon\varepsilon_l + (2 - c^2 - c)(\varepsilon_l - \varepsilon)]^2}{(1+c)[1 + (1-c)\varepsilon]} \quad \text{and} \quad \frac{\Pi_{22}}{H_{22}^m} = \frac{1}{H_{11}^m}, \quad \frac{\Pi_{33}}{H_{33}^m} = 0 \quad \text{(45)}
\]

Figure 4 shows plots of the electrostriction ratio \( \Pi_{11}/H_{11}^m \) for fibers with two different permittivities: \( \varepsilon_l = 4\varepsilon_0 \) and \( 10^4\varepsilon_0 \). The former value corresponds to the permittivity of Nylon, while the latter can be viewed as the permittivity of carbon. Both of these materials have been widely utilized as reinforcing fibers in elastomers (see, e.g., Refs. [16,21]). The results are plotted as functions of the volume fraction of fibers \( c \). In this and subsequent figures, the solid line, referred to as theory, stands for the ratio of electrostrictions based on the effective electromechanical constants (36). On the other hand, the dashed line stands for the response of dielectric elastomer composites with the polydisperse DCC microstructure discussed in Appendix A, while the solid circles correspond to the FE simulations of dielectric elastomer composites with the monodisperse microstructure discussed in Appendix B.

A key observation from Fig. 4 is that an infinitesimal addition of rigid fibers generates a 50% enhancement in electrostriction in the transverse direction to the fibers, namely, \( \Pi_{11} = 3/2H_{11}^m \) for \( c = 0^+ \). This abrupt enhancement is independent of the permittivity of the fibers, \( \varepsilon_l \), and solely due to their rigidity, which constrains the composite to deform only in the transverse \( \varepsilon_1-\varepsilon_2 \) plane (see Fig. 3). As the content of fibers increases, the electrostriction enhancement in the elastomer with the lower-permittivity fibers (\( \varepsilon_l = 4\varepsilon_0 \)) monotonically decreases, vanishing at a fiber volume fraction of about \( c = 0.2 \). The further increase in the content of fibers beyond that point leads to a reduction in electrostriction (\( \Pi_{11}/H_{11}^m < 1 \)). The electrostriction of the elastomer with the high-permittivity fibers (\( \varepsilon_l = 10^4\varepsilon_0 \)) exhibits similar trends, but its reduction occurs at a much slower rate. In fact, even at the relatively large volume fraction of \( c = 0.4 \), the electrostriction enhancement remains about 30%.

To illustrate the particular mechanical and anisotropic nature of the above-described enhancement, it proves helpful to consider the ratios \( \Pi_{11}/H_{11}^m \) and \( \Pi_{22}/H_{22}^m \), where

\[
\begin{align*}
\Pi_{11}^m &= \Pi_{11}^m = 1 - \frac{\varepsilon^2[(3 + c)\varepsilon + (1 - c)\varepsilon_l]^2}{2(1+c)[1 + (1-c)\varepsilon]} \quad \text{(47)}
\end{align*}
\]

Similar to Fig. 4, Fig. 5 shows plots of the ratio \( \Pi_{11}/H_{11}^m \) for fibers with the two different permittivities \( \varepsilon_l = 4\varepsilon_0 \) and \( 10^4\varepsilon_0 \) in terms of the volume fraction of fibers \( c \). Contrary to Fig. 4, as expected on physical grounds, an infinitesimal addition of rigid fibers does not yield any enhancement of the plane-strain electrostriction as \( \Pi_{11}/H_{11}^m = 1 \) for \( c = 0^+ \). Further addition of fibers leads to a significant reduction in the plane-strain electrostriction for the case of low-permittivity (\( \varepsilon_l = 4\varepsilon_0 \)) fibers, and to a tenuous reduction for the case of high-permittivity (\( \varepsilon_l = 10^4\varepsilon_0 \)) fibers, at least for the range of volume fractions considered. This behavior of a 2D isotropic distribution of rigid circular disks is analogous to the response of its 3D counterpart, an isotropic distribution of rigid spherical particles, presented in Ref. [2].

![Fig. 4 Electrostriction ratio \( \Pi_{11}/H_{11}^m \) of an incompressible \( (\lambda = \infty) \) dielectric elastomer with permittivity \( \varepsilon = 3.2\varepsilon_0 \) filled with a transversely isotropic distribution of rigid \( (\mu_l = \lambda_l = \infty) \) cylindrical fibers with circular cross section; see Fig. 3. Results are shown for fibers with permittivities \( \varepsilon_l = 4\varepsilon_0 \) and \( 10^4\varepsilon_0 \), as functions of the volume fraction of fibers \( c \). The solid line corresponds to the theoretical result (45). The dashed line corresponds to the response of a DCC assemblage of polydisperse fibers (see Appendix A), while the solid circles correspond to FE simulations of a dielectric elastomer composite with monodisperse fibers (see Appendix B).](http://appliedmechanics.asmedigitalcollection.asme.org/)

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These values are representative of silicone rubber.

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shear resistance, they are incompressible, composites with fibers that are liquid-like, in the sense that sufficiently far away from its percolation limit. shape and the size of the fibers, so long as the microstructure is “higher-order” microstructural details such as the cross-sectional SECTION, see Fig. 3. Results are shown for fibers with permittivities \( \varepsilon_1 = 4\varepsilon_0 \) and \( 10^5\varepsilon_0 \), as functions of the volume fraction of fibers \( c \). The solid line corresponds to the theoretical result (47). The dashed line corresponds to the response of a DCC assemblage of polydisperse fibers (see Appendix A), while the solid circles correspond to FE simulations of a dielectric elastomer composite with monodisperse fibers (see Appendix B).

Another key observation from Figs. 4 and 5 is the close agreement between the theoretical predictions (45), (47), and the DCC and FE results. This agreement among three different exact results for three different microstructures suggests that the electrostriction capacitivities of dielectric elastomers filled with transversely isotropic distributions of cylindrical fibers is fairly insensitive to “higher-order” microstructural details such as the cross-sectional shape and the size of the fibers, so long as the microstructure is sufficiently far away from its percolation limit.

### 4.2 Liquid-Like Fibers

Next, we consider dielectric elastomer composites with fibers that are liquid-like, in the sense that they are incompressible, \( \lambda_2 = \infty \), and exhibit vanishingly small shear resistance, \( \lambda_s = 0 \). Fibers with such properties could be made, for instance, by filling manufactured cylindrical cavities in the dielectric elastomer of interest with common fluids such as water or with eutectic alloys such as Galinstan [20,22,23]. As already alluded to in Ref. [2], an attractive feature of this type of dielectric elastomer composites is that increasing their content of fibers can increase their overall permittivity (if \( \varepsilon_s > \varepsilon \)) at the same time that it also increases their overall deformability (since \( \mu_s < \mu \)) and thus has the potential to bestow the resulting composites with exceptionally enhanced electrostriction capacitivities. The electrostriction components (43), when normalized by the matrix electrostriction components (44), reduce in this case to

\[
\frac{\Pi_{11}}{H_{11}} = 1 + \frac{\varepsilon(9 - \varepsilon)}{4(1 - \varepsilon)^2} + \frac{\varepsilon \varepsilon_s}{(1 - \varepsilon)^2} \times \frac{e + (3e^2 + 5)(e - e_s) - c(e_s + 8e\varepsilon_s)}{[(1 + c)e + (1 - c)e_s]^2},
\]

\[
\frac{\Pi_{22}}{H_{22}} = \frac{3}{2} \left( \frac{\Pi_{11}}{H_{11}} + \frac{\Pi_{33}}{H_{33}} \right),
\]

\[
\frac{\Pi_{33}}{H_{33}} = 1 + \frac{e[(5 - 3)e_s - (1 - c)e]}{(1 - c)[(1 + c)e + (1 - c)e_s]}.
\]

Figure 6 shows the results for the electrostriction ratios \( \Pi_{11}/H_{11} \) and \( \Pi_{33}/H_{33} \) for fibers with permittivity \( \varepsilon = 10^2\varepsilon_0 \), a value representative of the permittivity of water, as functions of the volume fraction of fibers \( c \). As expected, the enhancement in both the electrostriction ratios with the addition of such liquid-like fibers is very significant, exceeding 200% for fiber volume fractions \( c > 0.3 \). Consistent with the previous case of stiff fibers, the theoretical results (48) are seen to be in fairly good agreement with the DCC and FE results.

### 4.3 Vacuous Cylindrical Pores

As a final set of sample results, we consider the electrostriction of dielectric elastomer composites containing aligned cylindrical vacuous cavities or pores, \( \varepsilon_s = \mu_s = 0, \varepsilon_s = \varepsilon_0 \). For this choice of “fillers”, expressions (43), again, when normalized by (44), specialize to

\[
\frac{\Pi_{11}}{H_{11}} = 1 + \frac{(21 - c)\varepsilon \varepsilon_s}{4(1 - \varepsilon)^2} + \frac{\varepsilon \varepsilon_s}{(1 - \varepsilon)^2} \times \frac{e_0 + (10 + 9e)e + (8 - 9e)(e_0 - e)}{[(1 + c)e + (1 - c)e_0]^2},
\]

\[
\frac{\Pi_{22}}{H_{22}} = 1 + \frac{9 - 5e\varepsilon_s}{8(1 - \varepsilon)^2} + \frac{\varepsilon \varepsilon_s}{(1 - \varepsilon)^2} \times \frac{(1 - c)(3e + 7e_0) - (2 - c)(1 + 3e)e}{[(1 + c)e + (1 - c)e_0]^2},
\]

\[
\frac{\Pi_{33}}{H_{33}} = 1 - \frac{e[(1 - c)e - (3 - c)e_0]}{(1 - c)[(1 + c)e + (1 - c)e_0]}
\]

(49)

Figure 7 shows the plots of the three of these electrostriction ratios as functions of the volume fraction of pores \( c \). While the electrostriction ratio \( \Pi_{22}/H_{22} \) associated with the direction of the applied
Theoretical and experimental results are provided in the next section.

Theoretical and experimental results are provided in the next section.

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**Appendix A: The Overall Electromechanical Response of a Differential Coated Cylinder (DCC) Assemblage**

In this Appendix, we derive the analytical solution for the electromechanical response of a DCC assemblage in the limit of small deformations and moderate electric fields. The derivation follows the ideas introduced in Ref. [24], where the solution for the electromechanical response of a differential coated spherical assemblage was derived.

**We begin by introducing the notation**

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

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

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

**A.1 DCC Assemblages**

A coated cylinder assemblage is a two-phase particulate microstructure wherein aligned homothetic coated cylinders—comprising a cylindrical core of circular cross section made up of the fiber material that is surrounded by a cylindrical shell made up of the matrix material—of infinitely many sizes are assembled together to fill the entire domain \( \Omega \). This particular manner in which the coated cylinders are assembled is arbitrary. Assemblages where coated cylinders of comparable radius are placed far apart from each other and surrounded by coated cylinders of much smaller radius, in such a way that the microstructure is fractal-like comprising a hierarchy of well-separated coated cylinders, are referred to as DCC assemblages [26].

One of the two defining features of a DCC assemblage is that, by construction, any coated cylinder in the assemblage can be regarded to be surrounded by a homogeneous medium of infinite extent with the effective properties of the entire assemblage. The other defining feature being that the average response of any coated cylinder is the same as the average response of the entire assemblage (or, equivalently, that the so-called average “polarizability” of each coated cylinder vanishes); see Ref. [27] and Sec. 10.5 in the monograph by Milton [28]. These two defining features of such microstructures entail that the gradients of the tensor fields \( \Gamma \) and \( \gamma \) needed in expressions (A2) to compute the effective tensors \( L, \bar{e}, M \) are the same in each of the coated cylinders. This allows to simplify expressions (A2) to

**Fig. 7** Electrostriction ratios \( \frac{H_{33}}{H_{33}^m}, \frac{H_{22}}{H_{22}^m}, \frac{H_{11}}{H_{11}^m} \) of an incompressible \( (\lambda = \infty) \) dielectric elastomer with permittivity \( \varepsilon = 3.2 \varepsilon_0 \) containing a transversely isotropic distribution of vacuum \( (\rho_0 = \varepsilon_0 = \rho = 1) \) cylindrical pores with circular cross section, as functions of the volume fraction of pores \( c \). The solid line corresponds to the theoretical results (49). The dashed line corresponds to the response of a DCC assemblage of polydisperse cylindrical pores, while the solid circles correspond to the FE simulations of a dielectric elastomer composite with monodisperse cylindrical pores.
\[ L_{ijkl} = \frac{1}{|c|} \int_C L_{ijpq} \Gamma_{pklq} \, dX \]
\[ \bar{\varepsilon}_{ij} = \frac{1}{|c|} \int_C \varepsilon \epsilon_{ij} \, dX \]
\[ \bar{\rho}_{ijkl} = \frac{1}{|c|} \int_C \Gamma_{ijklpq} \rho_{pqrs} \epsilon_{rst} \, dX \]

where the integrals are now over the domain \( C \) occupied by a single coated cylinder, as opposed to over the entire domain \( \Omega \). Moreover, these gradients can be expediently computed by taking the domain \( \Omega \) in their defining boundary-value problems (A3) and (A4) to be an infinite body comprised of a single coated cylinder, occupying the domain \( C = \{ X : |X - (X \cdot N)| \leq 1 \} \) say, embedded in a homogeneous medium with the effective properties of the entire assembly. More specifically, when solving the boundary-value problem (A3) and (A4), it suffices to consider \( \Omega = \mathbb{R}^3 \), the local modulus of elasticity

\[
L = \begin{cases} 
(L^{(2)}) & \text{if } |X - (X \cdot N)| \leq R_e \\
(L^{(1)}) & \text{if } R_e \leq |X - (X \cdot N)| \leq 1 \\
\bar{L} & \text{if } |X - (X \cdot N)| \geq 1
\end{cases}
\] (A6)

the local permittivity tensor

\[
\epsilon = \begin{cases} 
(\epsilon^{(2)}) & \text{if } |X - (X \cdot N)| \leq R_e \\
(\epsilon^{(1)}) & \text{if } R_e \leq |X - (X \cdot N)| \leq 1 \\
\bar{\epsilon} & \text{if } |X - (X \cdot N)| \geq 1
\end{cases}
\] (A7)

and the local electrostrictive tensor

\[
M = \begin{cases} 
(M^{(2)}) & \text{if } |X - (X \cdot N)| \leq R_e \\
(M^{(1)}) & \text{if } R_e \leq |X - (X \cdot N)| \leq 1 \\
\bar{M} & \text{if } |X - (X \cdot N)| \geq 1
\end{cases}
\] (A8)

where \( R_e \) stands for the radius of the fiber within a coated cylinder of radius 1. For the class of dielectric elastomer composites of interest in Sec. 4, wherein the matrix and the fibers are isotropic ideal elastic dielectrics, the moduli of elasticity \( L^{(i)} \), permittivity tensors \( \epsilon^{(i)} \), and electrostrictive tensors \( M^{(i)} \) are given by expressions (13).

Given the local electromechanical tensors (A6)-(A8), the solutions to the PDEs (A3) and (A4) can be worked out in terms of solid harmonics (see, e.g., Ref. [29]). To simplify the calculations involved, it proves helpful to choose the direction of the fibers \( N \) to coincide with one of the laboratory axes. For \( N = e_3 \) (see Fig. 3), the 13 effective electromechanical constants (9) specialize to

\[
\bar{\sigma}_L = L_{1111} + L_{1222} = \frac{1}{|c|} \int_C L_{ijpq} \Gamma_{pklq} \rho_{ij} \, dX,
\]
\[
\bar{\rho}_{L} = L_{3333} = \frac{1}{|c|} \int_C L_{ijpq} \rho_{pklq} \epsilon_{ij} \, dX,
\]
\[
\bar{\epsilon}_L = 2L_{1212} = \frac{2}{|c|} \int_C L_{ijpq} \epsilon_{pklq} \, dX,
\]
\[
\bar{\rho}_L = 2L_{1313} = \frac{2}{|c|} \int_C L_{ijpq} \rho_{pklq} \epsilon_{ij} \, dX,
\]
\[
\bar{\epsilon}_L = L_{1133} = \frac{1}{|c|} \int_C L_{ijpq} \rho_{ij} \epsilon_{pklq} \, dX,
\]
\[
\bar{\rho}_L = \bar{\epsilon}_L = \bar{\rho}_L = \bar{\epsilon}_L = \frac{1}{|c|} \int_C \epsilon_{ij} \, dX,
\]

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

### A.2 The Solution for \( \Gamma \)

We begin by presenting the solution for the components \( \Gamma_{ij,k} \), which are needed in the computation of the effective elastic constant (A9), and the effective electrostrictive constant (A9). They read as

\[
\Gamma_{112,1} = \left[ \frac{g}{R} + 2h \right] X_1 X_2 \frac{h'}{R} X_2^3 X_2 \text{ for } 0 \leq R \leq R_e
\]
\[
\Gamma_{112,2} = \left[ \frac{g}{R} + 2h \right] X_1 X_2 \frac{h'}{R} X_1^3 X_2 \text{ for } R_e \leq R \leq 1
\]
\[
\Gamma_{112,3} = \frac{1}{2} \left[ \frac{g}{R} + h \right] R^2 + g + \frac{h'}{R} X_2^3 X_2 \text{ for } R \geq 1
\]
\[
\Gamma_{312,3} = \frac{h}{2} R^2 + B_2 R^2 + B_3 R^2 \text{ for } R \geq 1
\]

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

\[
g = \begin{cases} 
A_1 + A_2 R^2 & \text{if } 0 \leq R \leq R_e \\
B_1 + B_2 R^2 + B_3 R^2 & \text{if } R_e \leq R \leq 1
\end{cases}
\] (A11)

and

\[
h = \begin{cases} 
\frac{2(\lambda_2 + 3\mu)}{2\lambda_2 + 3\mu} A_3 & \text{if } 0 \leq R \leq R_e \\
-4B_2 \frac{2(\lambda_2 + 3\mu)}{2\lambda_2 + 3\mu} B_3 & \text{if } R_e \leq R \leq 1
\end{cases}
\] (A12)

In these expressions, use has been made of the notation \( g'(R) = \frac{dg}{dR} \), \( h'(R) = \frac{dh}{dR} \), and \( A_1, A_2, B_1, B_2, B_3 \) are constants that depend on the volume fraction of fibers \( c = R_e^2 \), on the effective properties of the matrix and the fibers, and on the effective elastic coefficients \( c_{ij} \) and \( \bar{c}_{ij} \) of the entire assembly. Because of their bulkiness, the explicit form of these constants is deferred to Sec. A.5.

We proceed with the solution for the combination of components \( \Gamma_{11,1} + \Gamma_{22,1} \). This combination is needed in the computation of the effective elastic constant (A9), and the effective electrostrictive constants (A9) and (A9). It can be written as follows:

\[
\bar{c}_L = M_{1111} + M_{2222} = \frac{1}{|c|} \int_C \left[ \Gamma_{p11,q} + \Gamma_{p22,q} \right] M_{pqrs} \epsilon_{ij} \, dX,
\]
\[
\bar{\rho}_M = M_{3333} = \frac{1}{|c|} \int_C \left[ \Gamma_{p33,q} \right] M_{pqrs} \epsilon_{ij} \, dX,
\]
\[
\bar{\epsilon}_M = 2M_{1212} = \frac{2}{|c|} \int_C \left[ \Gamma_{p11,q} + \Gamma_{p22,q} \right] M_{pqrs} \epsilon_{ij} \, dX,
\]
\[
\bar{\rho}_M = M_{1313} = \frac{1}{|c|} \int_C \left[ \Gamma_{p33,q} \right] M_{pqrs} \epsilon_{ij} \, dX,
\]
\[
\bar{\epsilon}_M = \bar{\rho}_M = \bar{\epsilon}_M = \bar{\rho}_M = \frac{1}{|c|} \int_C \epsilon_{ij} \, dX,
\]
\[
\begin{align*}
\Gamma_{111,1} + \Gamma_{122,1} &= \frac{d + e}{R^2} - \frac{2c}{R^2}X_1^2 \\
\Gamma_{211,2} + \Gamma_{222,2} &= \frac{d + e}{R^2} - \frac{2c}{R^2}X_2^2 \\
\Gamma_{111,2} + \Gamma_{122,2} &= \Gamma_{211,1} + \Gamma_{222,1} = -\frac{2c}{R^2}X_1X_2 \\
\Gamma_{111,3} + \Gamma_{122,3} &= \Gamma_{311,1} + \Gamma_{322,1} = 0 \\
\Gamma_{211,3} + \Gamma_{222,3} &= \Gamma_{311,2} + \Gamma_{322,2} = 0 \\
\Gamma_{311,3} + \Gamma_{322,3} &= 0
\end{align*}
\]

where \(d\) and \(e\) are given by
\[
d = \begin{cases} 
  d_e & \text{if } 0 \leq R \leq R_e \\
  d_m & \text{if } R_e \leq R \leq 1
\end{cases}
\]
and
\[
e = \begin{cases} 
  e_0 & \text{if } 0 \leq R \leq R_e \\
  e_m & \text{if } R_e \leq R \leq 1
\end{cases}
\]

with
\[
d_e = \frac{(\lambda - 2\mu)(\bar{e}_L + \bar{e}_L)}{(\lambda + \mu + \mu_e)(\bar{e}_L + \bar{e}_L)} - c(\lambda_e - \mu - \mu_e - \mu)(\bar{e}_L - 2\mu)
\]
\[
d_m = \frac{(\lambda + \mu + \mu_e)(\bar{e}_L + \bar{e}_L)}{(\lambda + \mu + \mu_e)(\bar{e}_L + \bar{e}_L)} - c(\lambda_e - \lambda + \mu_e - \mu)(\bar{e}_L - 2\mu)
\]
\[
e_m = \frac{-c(\lambda_e - \lambda + \mu_e - \mu)(\bar{e}_L + \bar{e}_L)}{(\lambda + \mu + \mu_e)(\bar{e}_L + \bar{e}_L)} - c(\lambda_e - \mu - \mu_e - \mu)(\bar{e}_L - 2\mu)
\]

Next, we record the solution for the components \(\Gamma_{113,j}\) required in the computation of the effective elastic constant (A9)_4 and the effective electrostrictive constant (A9)_11. These components read as
\[
\begin{align*}
\Gamma_{113,3} &= \Gamma_{313,1} = t + \frac{w}{R^2} \left[ 1 - 2X_1^2 \right] \\
\Gamma_{213,3} &= \Gamma_{313,2} = -\frac{2w}{R^2}X_1X_2 \\
\Gamma_{113,1} &= \Gamma_{313,2} = \Gamma_{113,3} = \Gamma_{213,1} = 0
\end{align*}
\]
where
\[
t = \begin{cases} 
  t_e & \text{if } 0 \leq R \leq R_e \\
  t_m & \text{if } R_e \leq R \leq 1
\end{cases}
\]
with
\[
t_e = \frac{2\mu \bar{e}_L}{(\mu + \mu_e)(\bar{e}_L + 2\mu) - c(\mu_e - \mu)(\bar{e}_L - 2\mu)}
\]
\[
t_m = \frac{2\mu \bar{e}_L}{(\mu + \mu_e)(\bar{e}_L + 2\mu) - c(\mu_e - \mu)(\bar{e}_L - 2\mu)}
\]
and
\[
u = \begin{cases} 
  v_e & \text{if } 0 \leq R \leq R_e \\
  v_m & \text{if } R_e \leq R \leq 1
\end{cases}
\]
with
\[
v_e = -\frac{2[\lambda_e + (1 - c)\mu] + (1 + c)\lambda_e\mu - (\lambda_e + 2\mu)\bar{e}_L + (1 - c)(\lambda_e - \lambda)\bar{e}_L}{2(\lambda_e + \mu + \mu_e)(\bar{e}_L + 2\lambda + \mu) - 2c(\lambda_e - \lambda + \mu_e - \mu)(\bar{e}_L - 2\mu)}
\]
\[
v_m = -\frac{2(\lambda_e + \mu + \mu_e)(\bar{e}_L - \lambda + \mu_e - \mu)(\bar{e}_L - 2\mu)}{2(\lambda_e + \mu + \mu_e)(\bar{e}_L + 2\lambda + \mu) - 2c(\lambda_e - \lambda + \mu_e - \mu)(\bar{e}_L - 2\mu)}
\]
\[
w_m = -\frac{2c(\lambda_e - \lambda + \mu_e - \mu)(\bar{e}_L + 2\lambda + \mu) - 2c(\lambda_e - \lambda + \mu_e - \mu)(\bar{e}_L - 2\mu)}{2(\lambda_e + \mu + \mu_e)(\bar{e}_L + 2\lambda + \mu) - 2c(\lambda_e - \lambda + \mu_e - \mu)(\bar{e}_L - 2\mu)}
\]

Finally, we present the solution for the components \(\Gamma_{333,j}\), which are needed in the computation of the effective elastic constants (A9)_2 and (A9)_9, as well as of the effective electrostrictive constants (A9)_9 and (A9)_12. They read as
\[
\begin{align*}
\Gamma_{333,1} &= v + \frac{w}{R^2} \left[ 1 - 2X_1^2 \right] \\
\Gamma_{233,2} &= v + \frac{w}{R^2} \left[ 1 - 2X_1^2 \right] \\
\Gamma_{333,2} &= \Gamma_{233,1} = -\frac{2w}{R^2}X_1X_2 \\
\Gamma_{333,3} &= \Gamma_{333,3} = \Gamma_{333,3} = 0
\end{align*}
\]
where
\[
v = \begin{cases} 
  v_e & \text{if } 0 \leq R \leq R_e \\
  v_m & \text{if } R_e \leq R \leq 1
\end{cases}
\]
and
\[
w = \begin{cases} 
  0 & \text{if } 0 \leq R \leq R_e \\
  w_m & \text{if } R_e \leq R \leq 1
\end{cases}
\]
A.3 The Solution for $\gamma$

Having established the solution for the required components of the gradient of $\Gamma$, we now turn to present the solution for the components of the gradient of $\gamma$, which are needed in the computation of the two effective permittivity coefficients (A9)$_6$ and (A9)$_7$, as well as of the six effective electrostrictive coefficients (A9)$_{8-13}$. They read as follows:

$$
\begin{align*}
\gamma_{1,1} &= -\left[ a + \frac{b}{R^2} \right] + \frac{2b}{R^2} X_1^2 \tag{A26} \\
\gamma_{2,2} &= -\left[ a + \frac{b}{R^2} \right] + \frac{2b}{R^2} X_2^2 \\
\gamma_{1,2} &= \gamma_{2,1} = \frac{2b}{R^2} X_1 X_2 \\
\gamma_{3,3} &= 1 \\
\gamma_{1,3} &= \gamma_{2,3} = \gamma_{3,1} = \gamma_{3,2} = 0
\end{align*}
$$

where

$$
a = \begin{cases} 
a_\varepsilon & \text{if } 0 \leq R \leq R_\varepsilon \\
am & \text{if } R_\varepsilon \leq R \leq 1 \end{cases} \tag{A27}
$$

with

$$
a_\varepsilon = \frac{-4\varepsilon_l}{(\varepsilon_l + e)(\varepsilon_l + e' - \varepsilon)(\varepsilon_l - e)} \tag{A28} \\
am = \frac{-2(\varepsilon_l + e)e_l}{(\varepsilon_l + e)(\varepsilon_l + e' - \varepsilon)(\varepsilon_l - e)} \tag{A29}
$$

A.4 The Effective Electromechanical Constants

We are now equipped to determine the thirteen effective constants that characterize the overall electromechanical response of the assembly of coated cylinders. The integrals over a single coated cylinder in Eq. (A9) can be readily carried out by making use of the explicit solutions (A10), (A13), (A17), (A22) for the combinations of components $\Gamma_{1112}$, $\Gamma_{111j} + \Gamma_{122j}$, $\Gamma_{1j1j}$, $\Gamma_{j3j}$ of the gradient of $\Gamma$, as well as the components (A26) of the gradient of $\gamma$. Expressions (A9)$_1$-$3$ yield a system of coupled polynomial equations for the five effective elastic constants $c_L$, $d_L$, $\varepsilon_L$, $\beta_L$, $\gamma_L$, from which a unique solution can be extracted based on physical arguments. Expression (A9)$_6$ also yields a polynomial equation for the effective permittivity coefficient $\varepsilon_l$ from which a unique solution can be deduced. On the other hand, expressions (A9)$_8$-$13$ render explicit solutions for the effective electrostrictive coefficient $e_l$ and the effective electrostrictive constants $e_M$, $\delta_M$, $\beta_M$, $\gamma_M$. After some algebraic manipulation, the result for all the 13 effective electromechanical constants can be written as

$$
c_L^{DCC} = 2\lambda + 2\mu + \frac{2c(\lambda + 2\mu)(\Delta\lambda + \Delta\mu)}{\lambda + 2\mu + (1 - c)(\Delta\lambda + \Delta\mu)}, \quad d_L^{DCC} = \lambda + 2\mu + c(\Delta\lambda + 2\Delta\mu) - \frac{c}{\lambda + 2\mu + (1 - c)(\Delta\lambda + \Delta\mu)}
$$

$$
e_L^{DCC} = \frac{1 + (1 - c)\Delta\lambda}{2\lambda + 2\mu + (1 - c)(\Delta\lambda + \Delta\mu)}, \quad \varepsilon_L^{DCC} = \frac{1 + (1 - c)\Delta\lambda}{2\lambda + 2\mu + (1 - c)(\Delta\lambda + \Delta\mu)}
$$

$$
e_M^{DCC} = 2A_1\varepsilon_L^2c_\varepsilon c_\varepsilon + \frac{3a_1^2c_\varepsilon^2(\varepsilon_l + \mu + \nu)X_1^2}{2\lambda + 3\mu} + 2a_1^2(1 - c)\varepsilon_L^2B_1 - 2b_1^2(1 - c)\varepsilon_L^2B_2 - 2b_1^2(1 - c)\varepsilon_L^2(1 - c)B_2
$$

$$
\beta_M^{DCC} = e + \frac{2c_\varepsilon}{2\varepsilon_l + (1 - c)\Delta\varepsilon}, \quad \gamma_M^{DCC} = \frac{\varepsilon}{2\varepsilon_l + (1 - c)\Delta\varepsilon} + \frac{c(1 - c)\Delta\lambda(2\varepsilon_l + \Delta\varepsilon)\Delta\lambda}{2\varepsilon_l + (1 - c)\Delta\varepsilon} \tag{A32}
$$

where the superscript “DCC” has been appended for clarity. In these expressions, it is recalled that $\Delta\lambda = \lambda - \lambda_e$, $\Delta\mu = \mu - \mu_e$, $\Delta\varepsilon = \varepsilon - \varepsilon_l$, the constants $a_\varepsilon$, $a_m$, $b_\varepsilon$ are given by expressions (A28), (A29), (A31), the constants $A_1$, $A_3$, $B_1$, $B_2$, $B_3$, $B_4$ are defined in Sec. A.5, and the final set of constants $q_1$, $q_2$, $q_3$, which depend explicitly on the volume fraction of fibers $c$ and the elastic properties of the matrix and the fibers, are given by expressions (A34) in Sec. A.6. A few comments are in order:

(1) With the exception of the effective elastic constant $e_L^{DCC}$ and the effective electrostrictive constant $\varepsilon_M^{DCC}$, all of the remaining 11 effective electromechanical constants (A32)
agree identically with the result (36) in the main body of the text. This agreement is admittedly remarkable since the two sets of results pertain to two different microstructures. These microstructures, however, do have the same one- and two-point correlations functions. Furthermore, these microstructures are similar in that the fibers in both of them can act as "neutral inclusions" under the same type of loading conditions (see Appendix B in Ref. [1]).

(2) We recall here that the five effective elastic constants $E_1^{DCC}$, $E_2^{DCC}$, $E_3^{DCC}$, $E_4^{DCC}$, $E_5^{DCC}$, can be recast into the five more conventional elastic parameters, $\mu_E$, $\nu_E$, $\kappa_E$, $\mu_F$, $\nu_F$, $\kappa_F$, $E_1$, $E_2$, $E_3$, $E_4$, $E_5$, utilized to characterize the elastic response of transversely isotropic materials, via relations (11). In this connection, it is not difficult to verify that the effective transverse shear modulus $\mu_E$ agrees identically with the result originally derived by Christensen and Lo [30], and later proved to be realizable by Avellaneda [26], for a DCC assembly. Further, the remaining four effective elastic constants $E_2^{DCC}$, $E_3^{DCC}$, $E_4^{DCC}$, $E_5^{DCC}$ agree identically with the classical results of Hashin and Rosen [25] and Hill [31] for arbitrary (not necessarily differential) coated cylinder assemblers. Similarly, the two effective permittivities $\varepsilon_E^{DCC}$ and $\varepsilon_F^{DCC}$ agree identically with the classical results (see Ref. [32] and references therein) for arbitrary (not necessarily differential) coated cylinder assemblers. By contrast, this is not true for the effective electrostrictive coefficient $\varepsilon_M^{DCC}$, which is only valid for DCC assemblers.

A.5 The Constants $A_1, A_3, B_1, B_2, B_3, B_4$

The six constants $A_1, A_3, B_1, B_2, B_3, B_4$ in the functions (A11) and (A12) are defined by the solution of the following system of six linear algebraic equations:

$$
\begin{align*}
E_1 &= A_1 + A_3 c - B_1 - B_2 c - B_3 c - B_4 c = 0, \\
E_2 &= A_4 \frac{2\lambda + 3\mu}{2\varepsilon^2} - B_1 - B_2 c - B_3 c - B_4 c = 0, \\
E_3 &= A_1 c^2 - B_1 c^2 - B_3 c^2 + B_4 c = 0, \\
E_4 &= A_4 c^2 - B_1 c^2 - B_2 c^2 + B_3 c^2 - B_4 c = 0, \\
E_5 &= B_1 \left(1 + \frac{4\mu}{\sigma_L} + \frac{2\mu}{\sigma_L} \right) + B_2 \left(1 - \frac{2\mu}{\sigma_L} \right) + B_3 \left(1 + \frac{2\mu}{\sigma_L} \right) + B_4 \left(1 - \frac{2\mu}{\sigma_L} \right) - \frac{2\lambda + 3\mu}{\sigma_L} = 0.
\end{align*}
$$

(33)

These equations stem from the continuity of the fields $\Gamma_{\varepsilon d}$ and $L_{\rho d}\Gamma_{e d}e_{d j}$ across the two material interfaces at $R = R_e$, 1, and from the boundary condition at infinity where $\Gamma_{\varepsilon d} = \delta_2 \delta_{3 d} X_3$. While it is a simple matter to compute the unique solution to this system of equations, we do not report it here because of the bulkiness of the final expressions.

A.6 Expressions for $q_1, q_2, q_3$

The expressions for $q_1, q_2, q_3$ in the result (A9) for the effective elastic constant $\delta_2^{DCC}$ are given by

$$
\begin{align*}
q_1 &= \left(\lambda + 3\mu\right)\tau_1 c^4 - 4\tau_2 c^3 + \left(\lambda + 3\mu\right)\tau_3 c^3, \\
q_2 &= \mu \tau_1 c^4 - 4\tau_2 c^3 + \tau_3 c^4, \\
q_3 &= \left(\lambda + \mu\right)\tau_1 c^4 - 4\tau_2 c^3 + \tau_3 c^4 - 2\mu c \left(\lambda + \mu\right) c^3 - 2\mu c^3. \\
\end{align*}
$$

(A34)

where

$$
\begin{align*}
\tau_1 &= \left(\mu - \mu\right)\left(\mu - \mu\right)\left(\mu - \mu\right)\left(\mu - \mu\right)\left(\mu - \mu\right), \\
\tau_2 &= -\lambda + 3\mu\left(\lambda + \mu\right) + \mu c, \\
\tau_3 &= \left(\lambda + \mu\right)\left(\lambda + \mu\right)\left(\lambda + \mu\right)\left(\lambda + \mu\right)\left(\lambda + \mu\right),
\end{align*}
$$

(A35)

Appendix B: FE Calculations of the Overall Elastic Dielectric Response of Elastomers Strengthened/Weakened by Aligned Cylindrical Fibers

In this Appendix, we provide details of the FE results presented in Sec. 4 for the overall electromechanical response of dielectric elastomers filled with a random distribution of aligned fibers with monodisperse circular cross section. We follow the formulation presented in Appendix A of Spinelli et al. [2], where the FE results for the electromechanical response of isotropic suspensions of spherical particles in rubber were established.

**Problem setting.** We recall that the general formulae (A2) presented in Appendix A for the effective electromechanical tensors $\mathbf{L}$, $\mathbf{e}$, $\mathbf{M}$ of dielectric elastomer composites with arbitrary heterogeneous local modulus of elasticity, $\mathbf{L} = \mathbf{L}(X)$, permittivity, $\epsilon = \epsilon(X)$, and electrostrictive tensor, $\mathbf{M} = \mathbf{M}(X)$, require solving the three-dimensional boundary-value problems (A3) and (A4) for the tensor fields $\mathbf{F}$ and $\gamma$. Now, for microstructures comprising aligned cylindrical fibers, the heterogeneity is two-dimensional. By choosing—as in the forgoing and without loss of generality—the direction of the fibers $\mathbf{N}$ to coincide with the laboratory axis $\mathbf{e}_1$ (see Fig. 3), the local electromechanical tensors for the problem of interest here read as

$$
\mathbf{L}(X) = \left[3\lambda(X_1, X_2) + 2\mu(X_1, X_2)\right]J + 2\mu(X_1, X_2)K, \\
\mathbf{e}(X) = e(X_1, X_2)I, \\
\mathbf{M}(X) = \epsilon(X_1, X_2) \left(\mathbf{K} - \frac{\mathbf{J}}{2}\right),
$$

(B1)

Because of the isotropy of these tensors and their independence of the $X_3$ coordinate, it follows that the three-dimensional boundary-
value problems (A3) and (A4) can actually be recast as two-dimensional boundary-value problems:

The isotropy of the local modulus of elasticity (B1) and its independence from $X_i$ imply that

$$
\Gamma_{q11,r} = \Gamma_{q11,l}(X_1, X_2), \quad \Gamma_{q22,r} = \Gamma_{q22,l}(X_1, X_2)
$$

$$
\Gamma_{q12,r} = \Gamma_{q12,l}(X_1, X_2), \quad \Gamma_{q21,r} = \Gamma_{q21,l}(X_1, X_2)
$$

$$
\Gamma_{q33,r} = \Gamma_{q33,l}(X_1, X_2)
$$

(B2)

and that $\Gamma_{q11}, \Gamma_{q22}, \Gamma_{q12}, \Gamma_{q21}, \Gamma_{q33}$ are solutions to

$$
[\tilde{\gamma}(X_1, X_2) \Gamma_{qij,s} \delta_{ij} + 2\mu(X_1, X_2) \Gamma_{qkl,s}]_{ij} = 0, \quad X \in \Omega
$$

with $\Gamma_{qkl} = \delta_{qk} X_l, \quad X \in \partial \Omega$

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

$$
\Gamma_{3q3,3} = \Gamma_{3q3,1}(X_1, X_2)
$$

$$
\Gamma_{3q1,3} = \Gamma_{3q1,2}(X_1, X_2)
$$

$$
\Gamma_{3q2,3} = \Gamma_{3q2,1}(X_1, X_2)
$$

(B4)

and $\Gamma_{3q3}$, $\Gamma_{3q3}$ are solutions to

$$
[p(X_1, X_2) \Gamma_{3q3,s}]_{ij} = 0, \quad X \in \Omega
$$

with $\Gamma_{3q3} = \delta_{3q} X_l, \quad X \in \partial \Omega$

where now the pair of indices $(kl)$ are to be evaluated as 13, 31, 23, 31. In expressions (B2)–(B5) and subsequently, Greek indices range from 1 to 2.

Similarly, the isotropy of the local permittivity (B1) and its independence from $X_i$ imply that

$$
\gamma_{q1} = \gamma_{q1}(X_1, X_2), \quad \gamma_{q2} = \gamma_{q2}(X_1, X_2), \quad \gamma_{q3} = 1
$$

(B6)

and $\gamma_q$ are solutions to

$$
[e(X_1, X_2) \gamma_{q,s}]_{ij} = 0, \quad X \in \Omega, \quad \text{with} \quad \gamma_q = X_q, \quad X \in \partial \Omega
$$

(B7)

It is plain that the boundary-value problems, (B3), (B5), (B7), do not admit explicit solutions other than for special cases (see, e.g., Appendix A). However, the underlying PDEs being second-order linear elliptic, it is straightforward to solve them numerically by means of the FE method, as outlined next.

**FE formulation.** The focus of Sec. 4 is on dielectric elastomers strengthened/weakened by transversely isotropic distributions of aligned cylindrical fibers with circular cross section. To approximate the transverse isotropy of this class of microstructures, and in view of the two dimensionality of the boundary-value problems (B3), (B5), (B7), we consider an infinite two-dimensional periodic medium described by the periodic repetition of a square unit cell, $S = [X : X_i \in [0, 1], \quad i = 1, 2], \quad n$ that comprises a large but finite number $N$ of randomly distributed circles. Previous results based on this approach (see, e.g., Ref. [33]) have indicated that $N = 60$ circles is, in general, sufficient to approximate the transverse isotropy of the microstructure. Figure 8 depicts representative examples of such unit cells with $N = 60$ circles for various volume fractions of fibers: (a) $c = 0.05$, (b) $c = 0.15$, and (c) $c = 0.25$.

For the above-described repeating unit cells $S$, the boundary value problems (B3), (B5), and (B7) specialize to

$$
[\tilde{\gamma}(X_1, X_2) \Gamma_{qij,s} \delta_{ij} + 2\mu(X_1, X_2) \Gamma_{qkl,s}]_{ij} = 0, \quad X \in S
$$

with

$$
\Gamma_{qkl}(1, X_2) - \Gamma_{qkl}(0, X_2) = \delta_{qk} \delta_{l1}, \quad X \in \partial S
$$

(B8)

and

$$
[p(X_1, X_2) \Gamma_{3q3,s}]_{ij} = 0, \quad X \in S
$$

with

$$
\Gamma_{3q3}(1, X_2) - \Gamma_{3q3}(0, X_2) = \delta_{3q} \delta_{l1}, \quad X \in \partial S
$$

(B9)

and

$$
[e(X_1, X_2) \gamma_{q,s}]_{ij} = 0, \quad X \in S
$$

with

$$
\gamma_q(1, X_2) - \gamma_q(0, X_2) = \delta_{q1}, \quad X \in \partial S
$$

(B10)

In the above expressions, $\partial S$ stands for the boundary of $S$ and it is recalled that the pair of indices $(kl)$ are to be evaluated as 11, 22, 12, 21, 33 in Eq. (B8) and as 13, 31, 23, 31 in Eq. (B9).

Fig. 8 Unit cells with $N = 60$ circles randomly distributed for three volume fractions of fibers: (a) $c = 0.05$, (b) $c = 0.15$, and (c) $c = 0.25$. Parts (d), (e), and (f) show three progressively refined meshes, with approximately (d) 8,000, (e) 40,000, and (f) 200,000 elements, of a unit cell containing a random distribution of $N = 60$ circles at volume fraction $c = 0.15$. 

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We carry out the discretization of these problems with help of the scripting and meshing capabilities of the FE package ABAQUS (see ABAQUS Version 6.11 Documentation, 2011 [34]). Hybrid isoparametric eight-node quadrilateral elements with linearly varying pressure were selected to solve the PDE (B8), while isoparametric eight-node quadrilateral elements were utilized for the PDEs (B9) and (B10). Figures 8(b)–8(d) display three meshes of increasing refinement for volume fraction of fibers $c = 0.15$. Mesh sensitivity analyses revealed that meshes containing about 200,000 elements (~ 600,000 nodes), such as the one shown in Fig. 8(f), are refined enough to deliver accurate results. The discretized equations resulting from Eqs. (B8), (B9), and (B10) are formulated and solved via a vectorized FE code written in the technical computing environment MATLAB (see MATLAB Version 8.3 Documentation, 2014 [35]). The computed FE solutions for the components of the tensor fields $\Gamma$ and $\gamma$ are then utilized to finally compute the three sought after effective electromechanical tensors via

\[
\begin{align*}
\delta_L &= \frac{\|\delta_{ij} L^{(E)}(\Gamma) + \delta_{ij} L^{(E)}(\gamma) + \delta_{ij} L^{(E)}(\psi) - \|L^{(E)}\|_\infty}{\|L^{(E)}\|_\infty} \\
\delta_c &= \frac{\|\delta_{ij} L^{(E)}(c - N \otimes N) + \delta_{ij} L^{(E)}(N \otimes N) - \delta_{ij} L^{(E)}\|_\infty}{\|L^{(E)}\|_\infty} \\
\delta_M &= \frac{\|\delta_{ij} L^{(E)}(M) + \delta_{ij} L^{(E)}(\psi) + \delta_{ij} L^{(E)}(\psi) - \|M^{(E)}\|_\infty}{\|M^{(E)}\|_\infty}
\end{align*}
\] (B12)

where the effective electromechanical coefficients $c_L$, $c_M$, $c_g$, $c_E$, $c_{\psi}$, $c_{\gamma}$, $c_{\psi}$, $c_{\gamma}$ are defined by the projections

\[
\begin{align*}
c_{\psi}^{(E)} &= \frac{1}{2} \left( - \delta_{ij} L^{(E)}(\psi) \delta_{ij} L^{(E)}(\psi) - \delta_{ij} L^{(E)}(\psi) \delta_{ij} L^{(E)}(\psi) \right) \\
c_{\gamma}^{(E)} &= \frac{1}{2} \left( - \delta_{ij} L^{(E)}(\gamma) \delta_{ij} L^{(E)}(\gamma) - \delta_{ij} L^{(E)}(\gamma) \delta_{ij} L^{(E)}(\gamma) \right) \\
c_{M}^{(E)} &= \frac{1}{2} \left( - \delta_{ij} L^{(E)}(M) \delta_{ij} L^{(E)}(M) - \delta_{ij} L^{(E)}(M) \delta_{ij} L^{(E)}(M) \right)
\end{align*}
\] (B13)

Here, we remark the fourth order tensors $E^{(i)}$ defined in Eq. (10) are to be evaluated with $N = \epsilon_1$. For the class of dielectric elastomer composites of interest in Sec. 4, wherein the matrix and particles are both isotropic elastic dielectrics, a perfectly transversely isotropic microstructure would result in measures $\delta_L = \delta_c = \delta_M = 0$. In this work, only realizations with

\[
\max\{\delta_L, \delta_c, \delta_M\} \leq 10^{-2}
\] (B14)

are admitted as approximately transversely isotropic.

We conclude by noting that all the FE results presented in Sec. 4 correspond to the average of three different realizations (all satisfying the above definition of approximate isotropy). The pertinent components of the electrostrictive $\Pi$ displayed in the plots in that section are based on the formulae

\[
\begin{align*}
\Pi_{11} &= -\left( -\delta_{ij} L^{(E)}(\psi) - \delta_{ij} L^{(E)}(\psi) - \delta_{ij} L^{(E)}(\psi) \right) \frac{E^2}{2} \\
\Pi_{22} &= -\left( -\delta_{ij} L^{(E)}(\psi) - \delta_{ij} L^{(E)}(\psi) - \delta_{ij} L^{(E)}(\psi) \right) \frac{E^2}{2} \\
\Pi_{33} &= -\left( -\delta_{ij} L^{(E)}(\psi) - \delta_{ij} L^{(E)}(\psi) - \delta_{ij} L^{(E)}(\psi) \right) \frac{E^2}{2}
\end{align*}
\] (B15)

References


[34] ABAQUS Version 6.11 Documentation, 2011, Dassault Systemes Simulia Corp., Providence, RI.