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The overall elastic dielectric properties of a suspension of spherical particles in rubber: An exact explicit solution in the small-deformation limit

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A solution is constructed for the homogenization problem of the elastic dielectric response of rubber filled with a random isotropic distribution of polydisperse spherical particles in the classical limit of small deformations and moderate electric fields. In this limit, the overall elastic dielectric response is characterized by five (two elastic, one dielectric, and two electrostrictive) effective constants. Explicit formulas are derived for these constants directly in terms of the corresponding constants describing the elastic dielectric response of the underlying rubber and the filler particles, as well as the concentration of particles. By means of comparisons with finite-element simulations, these formulas are shown to also be applicable to isotropic suspensions of monodisperse spherical particles, provided that the particle concentration is sufficiently away from percolation. With the aim of gaining physical insight into the extreme enhancement in electrostrictive properties displayed by emerging dielectric elastomer composites, specific results are examined for the case of suspensions wherein the rubber is incompressible and the particles are mechanically rigid and of infinite permittivity. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4897199]

I. INTRODUCTION

Modern advances in organic materials have recently led to the rediscovery of dielectric elastomers as a promising material choice to enable a wide host of new technologies.[1,2] Yet, because these soft low-permittivity materials require extremely high electric fields (>100 MV/m) to be actuated, their use in applications has remained limited. Over the past decade, experimental investigations have revealed that a possible solution to circumvent this limitation and to generally enhance the electrostrictive properties of dielectric elastomers is to make composite materials, essentially by adding high-permittivity or (semi-)conducting particles to the dielectric elastomers.[3–10] The precise nature behind the enhancement in electrostrictive properties generated by the addition of particles remains unresolved, but a number of microscopic mechanisms have been identified, including the nonlinear electromechanical nature of elastomers (which heightens the role of the fluctuations of the electric field)[11,12] and the presence of high-dielectric interphases and/or interphasial free charges.[13,14]

In this paper, we seek to gain precise quantitative insight into the enhancement that the nonlinear electromechanical nature of elastomers bestows on dielectric elastomer composites. To this end, we construct a rigorous solution for the homogenization problem of the elastic dielectric[15] response of rubber filled with a random isotropic distribution of firmly bonded polydisperse spherical particles. This choice of idealized microstructure, an isotropic suspension of polydisperse spherical particles, is thought to be a good approximation of the real microstructures investigated in many of the aforementioned experimental studies, especially those dealing with small concentration of particles where a good dispersion of roughly spherical particles was demonstrated. We restrict our analysis to the classical limit of small deformations and moderate electric fields.[12,16,17]

II. THE PROBLEM

Consider a random isotropic distribution of spherical particles that are firmly bonded to a rubber matrix. The domain occupied by this two-phase particulate composite material, or suspension, in its (undeformed and stress-free) ground state is denoted by Ω. The radii of the particles are taken to be much smaller than the length scale of Ω. Both, the rubber ($\mathrm{r}=1$) and the particles ($\mathrm{r}=2$) are elastic dielectrics whose properties are characterized by "total" free energies $W^{(\mathrm{r})}$ that are objective, isotropic functions of the deformation gradient tensor $\mathbf{F}$ and objective, isotropic, and even[18] functions of the Lagrangian electric field $\mathbf{E}$ so that $W^{(\mathrm{r})}(\mathbf{QFK},\mathbf{EK}) = W^{(\mathrm{r})}(\mathbf{F},-\mathbf{E}) = W^{(\mathrm{r})}(\mathbf{F},\mathbf{E})$ for all $\mathbf{Q}$, $\mathbf{K} \in Orth+$. At each material point $\mathbf{X} \in \Omega$, the first Piola-Kirchhoff stress tensor $\mathbf{S}$ and the Lagrangian electric displacement field $\mathbf{D}$ are hence given by[19,20]

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

with

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

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

Macroscopically, at the length scale of Ω, the overall electromechanical response for the above-defined composite

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material is characterized by the relation between the volume averages of the first Piola-Kirchhoff stress $\bar{\mathbf{S}} \doteq |\Omega|^{-1}\int_\Omega \mathbf{S}(\mathbf{X})d\mathbf{X}$ and electric displacement $\bar{\mathbf{D}} \doteq |\Omega|^{-1}\int_\Omega \mathbf{D}(\mathbf{X})d\mathbf{X}$ and the volume averages of the deformation gradient $\bar{\mathbf{F}} \doteq |\Omega|^{-1}\int_\Omega \mathbf{F}(\mathbf{X})d\mathbf{X}$ and electric field $\bar{\mathbf{E}} \doteq |\Omega|^{-1}\int_\Omega \mathbf{E}(\mathbf{X})d\mathbf{X}$ over the undeformed configuration Ω when the composite is subjected to affine boundary conditions. Such a relation can be compactly written as[21]

$$\bar{\mathbf{S}} = \frac{\partial \bar{W}}{\partial \bar{\mathbf{F}}}\left(\bar{\mathbf{F}}, \bar{\mathbf{E}}\right) \quad \text{and} \quad \bar{\mathbf{D}} = -\frac{\partial \bar{W}}{\partial \bar{\mathbf{E}}}\left(\bar{\mathbf{F}}, \bar{\mathbf{E}}\right), \tag{3}$$

where

$$\bar{W}\left(\bar{\mathbf{F}}, \bar{\mathbf{E}}\right) = \min_{\mathbf{F}\in\mathcal{K}} \max_{\mathbf{E}\in\mathcal{E}} \frac{1}{|\Omega|}\int_\Omega W(\mathbf{X}, \mathbf{F}, \mathbf{E})d\mathbf{X}. \tag{4}$$

In this last expression, $\mathcal{K}$, $\mathcal{E}$ denote sufficiently large sets of admissible deformation gradients $\mathbf{F}$ and curl-free electric fields $\mathbf{E}$ with prescribed volume averages $\bar{\mathbf{F}}$ and $\bar{\mathbf{E}}$. Physically, the so-called effective free energy function $\bar{W}$ corresponds to the total electroelastic free energy (per unit undeformed volume) of the composite. In the present context, much like its local counterpart (2), the effective free energy function $\bar{W}$ is objective and isotropic in $\bar{\mathbf{F}}$ and objective, isotropic, and even in $\bar{\mathbf{E}}$, namely, $\bar{W}(\bar{\mathbf{Q}}\,\bar{\mathbf{F}}\,\bar{\mathbf{K}}, \bar{\mathbf{E}}\,\bar{\mathbf{K}}) = \bar{W}(\bar{\mathbf{F}}, -\bar{\mathbf{E}}) = \bar{W}(\bar{\mathbf{F}}, \bar{\mathbf{E}})$ for all $\bar{\mathbf{Q}}, \bar{\mathbf{K}} \in Orth+$.

A. The classical limit of small deformations and moderate electric fields

The foregoing formulation is valid for arbitrarily large deformations and arbitrarily large electric fields. The focus of this paper is on the classical limit of *small macroscopic deformations* and *moderate macroscopic electric fields*.[12,16,17] More precisely, defining ζ as a vanishingly small parameter, the deformation measure $\bar{\mathbf{H}} \doteq \bar{\mathbf{F}} - \mathbf{I}$, with $\mathbf{I}$ denoting the identity in the space of second-order tensors, is assumed to be $O(\zeta)$ while the electric field $\bar{\mathbf{E}}$ is assumed to be $O(\zeta^{1/2})$. In this limit, the effective free energy function (4) takes the asymptotic form

$$\begin{aligned}\bar{W}\left(\bar{\mathbf{F}}, \bar{\mathbf{E}}\right) = {} & \frac{1}{2}\bar{H}_{ij}\tilde{L}_{ijkl}\bar{H}_{kl} - \frac{1}{2}\bar{E}_i\tilde{\epsilon}_{ij}\bar{E}_j + \bar{H}_{ij}\tilde{M}_{ijkl}\bar{E}_k\bar{E}_l \\ & - \bar{E}_i\bar{E}_j\tilde{\tau}_{ijkl}\bar{E}_k\bar{E}_l + O\left(\zeta^3\right),\end{aligned} \tag{5}$$

where $\tilde{\mathbf{L}}$ stands for the effective modulus of elasticity, $\tilde{\boldsymbol{\epsilon}}$ denotes the effective permittivity, $\tilde{\mathbf{M}}$ is the effective electrostrictive tensor, and $\tilde{\boldsymbol{\tau}}$ represents the effective permittivity of second order. The corresponding relations for the macroscopic stress and electric displacement (3) reduce to

$$\bar{S}_{ij} = \frac{\partial \bar{W}}{\partial \bar{F}_{ij}}\left(\bar{\mathbf{F}}, \bar{\mathbf{E}}\right) = \tilde{L}_{ijkl}\bar{H}_{kl} + \tilde{M}_{ijkl}\bar{E}_k\bar{E}_l + O\left(\zeta^2\right) \tag{6}$$

and

$$\bar{D}_i = -\frac{\partial \bar{W}}{\partial \bar{E}_i}\left(\bar{\mathbf{F}}, \bar{\mathbf{E}}\right) = \tilde{\epsilon}_{ij}\bar{E}_j + O\left(\zeta^{3/2}\right), \tag{7}$$

to leading order. It is important to recognize that the permittivity of second order $\tilde{\boldsymbol{\tau}}$ does *not* enter in either expression (6) or (7). That is, in this classical limit of small macroscopic deformations and moderate macroscopic electric fields, the overall electromechanical response of the suspension is characterized by three effective tensors: the fourth-order tensor $\tilde{\mathbf{L}}$ describing its elasticity, the second-order tensor $\tilde{\boldsymbol{\epsilon}}$ describing its permittivity, and the fourth-order $\tilde{\mathbf{M}}$ tensor describing its electrostrictive response.

Remarkably, in spite of the inherent coupling and nonlinearity of the problem, it is possible to write formulae for the effective electromechanical tensors $\tilde{\mathbf{L}}$, $\tilde{\boldsymbol{\epsilon}}$, $\tilde{\mathbf{M}}$ solely in terms of a purely elastic problem and an uncoupled purely dielectric problem.[12] With help of the notation $\mathbf{L} = [1-\theta(\mathbf{X})]\mathbf{L}^{(1)} + \theta(\mathbf{X})\mathbf{L}^{(2)}$, $\boldsymbol{\epsilon} = [1-\theta(\mathbf{X})]\boldsymbol{\epsilon}^{(1)} + \theta(\mathbf{X})\boldsymbol{\epsilon}^{(2)}$, $\mathbf{M} = [1-\theta(\mathbf{X})]\mathbf{M}^{(1)} + \theta(\mathbf{X})\mathbf{M}^{(2)}$, where $\mathbf{L}^{(\mathtt{r})} \doteq \partial^2 W^{(\mathtt{r})}(\mathbf{I},\mathbf{0})/\partial\mathbf{F}^2$, $\boldsymbol{\epsilon}^{(\mathtt{r})} \doteq \partial^2 W^{(\mathtt{r})}(\mathbf{I},\mathbf{0})/\partial\mathbf{E}^2$, $\mathbf{M}^{(\mathtt{r})} \doteq \partial^3 W^{(\mathtt{r})}(\mathbf{I},\mathbf{0})/\partial\mathbf{F}\partial\mathbf{E}^2$ $(\mathtt{r} = 1,2)$, for the local elastic modulus $\mathbf{L}$, local permittivity tensor $\boldsymbol{\epsilon}$, and local electrostrictive tensor $\mathbf{M}$ of the suspension, these formulae read as[12]

$$\tilde{L}_{ijkl} = \frac{1}{|\Omega|}\int_\Omega L_{ijrs}\Gamma_{rkl,s}\,d\mathbf{X}, \tag{8}$$

$$\tilde{\epsilon}_{ij} = \frac{1}{|\Omega|}\int_\Omega \epsilon_{is}\gamma_{s,j}\,d\mathbf{X}, \tag{9}$$

$$\tilde{M}_{ijkl} = \frac{1}{|\Omega|}\int_\Omega \Gamma_{rij,s}M_{rsuv}\gamma_{u,k}\gamma_{v,l}\,d\mathbf{X}, \tag{10}$$

where the tensor fields $\boldsymbol{\Gamma}$ and γ are defined implicitly as the solutions to the following linear *uncoupled* boundary value problems:

$$\left[L_{ijrs}\Gamma_{rkl,s}\right]_{,j} = 0, \mathbf{X}\in\Omega \quad \text{with} \quad \Gamma_{ikl} = \delta_{ik}\delta_{jl}X_j,\ \mathbf{X}\in\partial\Omega \tag{11}$$

and

$$\left[\epsilon_{is}\gamma_{s,j}\right]_{,i} = 0, \mathbf{X}\in\Omega \quad \text{with} \quad \gamma_i = \delta_{ij}X_j,\ \mathbf{X}\in\partial\Omega. \tag{12}$$

Here and subsequently, the notation, i represents partial differentiation with respect to the material point coordinate X_i, δ_{ij} denotes the Kronecker delta, and $\partial\Omega$ stands for the boundary of the domain Ω. It is apparent from formula (10) that large spatial fluctuations of the field $\boldsymbol{\gamma}$ would lead to an enhancement of the electrostrictive properties of the suspension. As outlined in the Introduction, a goal of this paper is to quantify how much of such an enhancement, if any, actually develops for the case of isotropic suspensions of spherical particles.

For later convenience, it is appropriate to remark that because of the constitutive isotropy of the rubber and the particles, the tensors $\mathbf{L}^{(\mathtt{r})}$, $\boldsymbol{\epsilon}^{(\mathtt{r})}$, $\mathbf{M}^{(\mathtt{r})}$ are of the form

$$\mathbf{L}^{(1)} = 2\mu\mathcal{K} + 3\kappa\mathcal{J}, \quad \boldsymbol{\epsilon}^{(1)} = \varepsilon\mathbf{I}, \quad \mathbf{M}^{(1)} = m_K\mathcal{K} + m_J\mathcal{J} \tag{13}$$

and

$$\mathbf{L}^{(2)} = 2\mu_{\mathrm{p}}\mathcal{K} + 3\kappa_{\mathrm{p}}\mathcal{J}, \quad \boldsymbol{\epsilon}^{(2)} = \varepsilon_{\mathrm{p}}\mathbf{I}, \quad \mathbf{M}^{(2)} = m_{K\mathrm{p}}\mathcal{K} + m_{J\mathrm{p}}\mathcal{J}. \tag{14}$$

In these expressions, μ, μ_{p}, κ, κ_{p}, ε, ε_{p}, m_K, $m_{K\mathrm{p}}$, m_J, $m_{J\mathrm{p}}$ stand for, respectively, the shear modulus, bulk modulus, permittivity, and electrostrictive coefficients of the rubber and the particles, and $\mathcal{K}$, $\mathcal{J}$ are two orthogonal projection tensors given by

$$\mathcal{K}_{ijkl}=\frac{1}{2}\left[\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-\frac{2}{3}\delta_{ij}\delta_{kl}\right],\quad \mathcal{J}_{ijkl}=\frac{1}{3}\delta_{ij}\delta_{kl}. \tag{15}$$

Moreover, because of the assumed overall (geometric and constitutive) isotropy of the suspension, the effective tensors (8)–(10) are also of the form

$$\tilde{\mathbf{L}}=2\tilde{\mu}\mathcal{K}+3\tilde{\kappa}\mathcal{J},\quad \tilde{\boldsymbol{\epsilon}}=\tilde{\varepsilon}\mathbf{I},\quad \tilde{\mathbf{M}}=\tilde{m}_K\mathcal{K}+\tilde{m}_J\mathcal{J}. \tag{16}$$

In turn, the overall electromechanical constitutive relations (6) and (7) read in this case simply as

$$\begin{aligned}\bar{\mathbf{S}}&=\tilde{\mathbf{L}}\,\bar{\mathbf{H}}+\tilde{\mathbf{M}}\,\bar{\mathbf{E}}\otimes\bar{\mathbf{E}}\\&=\tilde{\mu}\left[\bar{\mathbf{H}}+\bar{\mathbf{H}}^T-\frac{2}{3}(\mathrm{tr}\bar{\mathbf{H}})\mathbf{I}\right]+\tilde{\kappa}(\mathrm{tr}\bar{\mathbf{H}})\mathbf{I}\\&\quad+\tilde{m}_K\left[\bar{\mathbf{E}}\otimes\bar{\mathbf{E}}-\frac{1}{3}(\bar{\mathbf{E}}\cdot\bar{\mathbf{E}})\mathbf{I}\right]+\frac{\tilde{m}_J}{3}(\bar{\mathbf{E}}\cdot\bar{\mathbf{E}})\mathbf{I}\end{aligned} \tag{17}$$

and

$$\bar{\mathbf{D}}=\tilde{\epsilon}\,\bar{\mathbf{E}}=\tilde{\varepsilon}\,\bar{\mathbf{E}} \tag{18}$$

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

1. Electrostriction

In the absence of applied stresses when $\bar{\mathbf{S}}=\mathbf{0}$, it follows from the overall constitutive relation (17) that

$$\bar{\mathbf{H}}=-\frac{\tilde{m}_K}{2\tilde{\mu}}\bar{\mathbf{E}}\otimes\bar{\mathbf{E}}+\left[\frac{\tilde{m}_K}{6\tilde{\mu}}-\frac{\tilde{m}_J}{9\tilde{\kappa}}\right](\bar{\mathbf{E}}\cdot\bar{\mathbf{E}})\,\mathbf{I}. \tag{19}$$

The deformation measure (19) is referred to as the *electrostriction* that the suspension undergoes when it is subjected to a macroscopic electric field $\bar{\mathbf{E}}$.

For later reference, we note that in experiments, the electrostriction of isotropic deformable dielectrics, such as the class of suspensions of interest here, is commonly probed by sandwiching a thin layer of the material in between two compliant electrodes connected to a battery. In such a configuration, the resulting macroscopic stress is indeed roughly zero everywhere (inside the material as well as in the surrounding space), while the macroscopic electric field is roughly uniform and *uniaxial* within the material and zero outside of it. Upon denoting the Cartesian basis vectors of the laboratory axes by $\{\mathbf{e}_i\}$ $(i=1,2,3)$ and applying a uniaxial electric field of the form

$$\bar{\mathbf{E}}=\bar{E}\mathbf{e}_3 \tag{20}$$

with $\bar{E}=\Phi/L_3$, where Φ denotes the voltage applied between the electrodes and L_3 stands for the initial thickness of the layer of material, the deformation measure (19) takes the diagonal form

$$\bar{\mathbf{H}}=\bar{H}_{11}\mathbf{e}_1\otimes\mathbf{e}_1+\bar{H}_{22}\mathbf{e}_2\otimes\mathbf{e}_2+\bar{H}_{33}\mathbf{e}_3\otimes\mathbf{e}_3, \tag{21}$$

where the electrostriction transverse to the applied electric field is given by

$$\bar{H}_{11}=\bar{H}_{22}=\left(\frac{\tilde{m}_K}{6\tilde{\mu}}-\frac{\tilde{m}_J}{9\tilde{\kappa}}\right)\bar{E}^2, \tag{22}$$

while the electrostriction in the direction of the applied field is given by

$$\bar{H}_{33}=-\left(\frac{\tilde{m}_K}{3\tilde{\mu}}+\frac{\tilde{m}_J}{9\tilde{\kappa}}\right)\bar{E}^2. \tag{23}$$

III. THE SOLUTION FOR A SPECIAL CLASS OF SUSPENSIONS OF SPHERICAL PARTICLES: A DIFFERENTIAL COATED SPHERE ASSEMBLAGE

The boundary value problems (11) and (12) defining the tensors $\boldsymbol{\Gamma}$ and $\boldsymbol{\gamma}$ needed in the computation of the effective electromechanical tensors (8)–(10) are, in general, difficult to solve. This is because the indicator function $\theta(\mathbf{X})$ is discontinuous and highly oscillating. In this section, we consider a special class of suspensions of spherical particles (i.e., a special class of indicator functions $\theta(\mathbf{X})$) that allow for analytical solutions: *a differential coated sphere assemblage*.

We recall that a coated sphere assemblage[22] is a two-phase particulate microstructure wherein homothetic coated spheres—comprising a spherical core made up of the particle material that is surrounded by a spherical shell made up of the matrix material—of infinitely many sizes are assembled together to fill the entire domain Ω occupied by the composite. The particular manner in which the coated spheres are assembled is arbitrary. For assemblages wherein coated spheres of comparable size are placed far apart from each other and surrounded by coated spheres of much smaller size, in such a way that the microstructure is fractal-like comprising a hierarchy of well-separated coated spheres of infinitely many sizes, it is possible to construct analytical solutions for the corresponding boundary value problems (11) and (12). Following the terminology of Avellaneda[23] (see also the prior work of Milton[24] and Section 10.5 in Milton[25]), we refer to such assemblages as differential coated sphere assemblages. One of two defining features of such microstructures is that, by construction, any coated sphere in the assemblage can be regarded to be surrounded by a homogeneous medium of infinite extent with the effective properties of the entire assemblage. The other defining feature being that the average response of any coated sphere is the same as the average response of the entire assemblage[24,25] (or, equivalently, that the so-called average "polarizability" of each coated sphere vanishes[24]).

For the elastostatics and electrostatics problems (11) and (12) of interest here, the above-outlined features of a differential coated sphere assemblage entail that the gradients of the fields $\boldsymbol{\Gamma}$ and $\boldsymbol{\gamma}$ are the same in each one of the coated spheres and, therefore, that the formulae (8)–(10) for the effective electromechanical tensors $\tilde{\mathbf{L}}$, $\tilde{\boldsymbol{\epsilon}}$, $\tilde{\mathbf{M}}$ reduce to

$$\tilde{L}_{ijkl} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} L_{ijrs}\Gamma_{rkl,s}\,\mathrm{d}\mathbf{X}, \tag{24}$$

$$\tilde{\epsilon}_{ij} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} \epsilon_{is}\gamma_{s,j}\,\mathrm{d}\mathbf{X}, \tag{25}$$

$$\tilde{M}_{ijkl} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} \Gamma_{rij,s}M_{rsuv}\gamma_{u,k}\gamma_{v,l}\,\mathrm{d}\mathbf{X}, \tag{26}$$

where the integrals are now over *a single coated sphere*, denoted as $\mathcal{B}$, as opposed to over the entire assemblage Ω. Moreover, the gradients of the fields $\mathbf{\Gamma}$ and $\boldsymbol{\gamma}$ needed in (24)–(26) can be expediently computed by taking the domain Ω in the boundary value problems (11) and (12) to be an infinite body comprised of a single coated sphere, occupying the domain $\mathcal{B} = \{\mathbf{X}: \ |\mathbf{X}| \le 1\}$ say, embedded in a homogeneous medium with the effective properties of the entire assemblage. Namely, for the elastostatics problem (11), it suffices to consider $\Omega = \mathbb{R}^3$ and the local modulus of elasticity

$$\mathbf{L} = \begin{cases} \mathbf{L}^{(2)} = 2\mu_{\mathrm{p}}\mathcal{K} + 3\kappa_{\mathrm{p}}\mathcal{J} & \text{if} \quad |\mathbf{X}| \le R_{\mathrm{p}} \\ \mathbf{L}^{(1)} = 2\mu\mathcal{K} + 3\kappa\mathcal{J} & \text{if} \quad R_{\mathrm{p}} \le |\mathbf{X}| \le 1, \\ \tilde{\mathbf{L}} = 2\tilde{\mu}\mathcal{K} + 3\tilde{\kappa}\mathcal{J} & \text{if} \quad |\mathbf{X}| \ge 1 \end{cases} \tag{27}$$

while for the electrostatics problem (12), it suffices to consider $\Omega = \mathbb{R}^3$ and the local permittivity

$$\boldsymbol{\epsilon} = \begin{cases} \boldsymbol{\epsilon}^{(2)} = \varepsilon_{\mathrm{p}}\mathbf{I} & \text{if} \quad |\mathbf{X}| \le R_{\mathrm{p}} \\ \boldsymbol{\epsilon}^{(1)} = \varepsilon\mathbf{I} & \text{if} \quad R_{\mathrm{p}} \le |\mathbf{X}| \le 1, \\ \tilde{\boldsymbol{\epsilon}} = \tilde{\varepsilon}\mathbf{I} & \text{if} \quad |\mathbf{X}| \ge 1 \end{cases} \tag{28}$$

where R_{p} stands for the radius of the particle within the coated sphere of unit radius.

Given (27) and (28), the solutions to the boundary value problems (11) and (12) can be readily worked out in terms of solid harmonics. Before proceeding with the presentation of the solutions, we remark that

$$\tilde{\mu} = \tilde{L}_{1212} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} L_{12rs}\Gamma_{r12,s}\mathrm{d}\mathbf{X}, \tag{29}$$

$$\tilde{\kappa} = \frac{1}{9}\tilde{L}_{iijj} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} \frac{1}{9}L_{iirs}\Gamma_{rjj,s}\mathrm{d}\mathbf{X}, \tag{30}$$

$$\tilde{\varepsilon} = \frac{1}{3}\tilde{\epsilon}_{ii} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} \frac{1}{3}\varepsilon\gamma_{i,i}\mathrm{d}\mathbf{X}, \tag{31}$$

$$\tilde{m}_K = 2\tilde{M}_{1212} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} 2\Gamma_{r12,s}M_{rsuv}\gamma_{u,1}\gamma_{v,2}\mathrm{d}\mathbf{X}, \tag{32}$$

$$\tilde{m}_J = \frac{1}{3}\tilde{M}_{iijj} = \frac{1}{|\mathcal{B}|}\int_{\mathcal{B}} \frac{1}{3}\Gamma_{rii,s}M_{rsuv}\gamma_{u,j}\gamma_{v,j}\mathrm{d}\mathbf{X}. \tag{33}$$

That is, by virtue of the isotropic form (16) of the effective tensors $\tilde{\mathbf{L}}$, $\tilde{\boldsymbol{\epsilon}}$, $\tilde{\mathbf{M}}$, only certain combinations of the components of the gradients of the fields $\mathbf{\Gamma}$ and $\boldsymbol{\gamma}$ are needed for their computation. In the sequel, for conciseness, we report solutions for such combinations.

A. The solution for Γ

We begin by presenting the solution for the components $\Gamma_{i12,j}$, which are needed in the computation of the effective shear modulus (29) and the effective electrostrictive coefficient (32). Physically, such components are associated with the elastic response of the suspension under simple shear loading. As alluded to above, the solution can be easily constructed in terms of solid harmonics (see, e.g., Chapter XI in Love[26]). It reads as follows:

$$\begin{aligned}
\Gamma_{112,1} &= \left[\frac{f'}{R} + 2g\right]X_1X_2 + \frac{g'}{R}X_1^3X_2, \\
\Gamma_{212,2} &= \left[\frac{f'}{R} + 2g\right]X_1X_2 + \frac{g'}{R}X_1X_2^3, \\
\Gamma_{312,3} &= gX_1X_2 + \frac{g'}{R}X_1X_2X_3^2, \\
\Gamma_{112,2} &= \Gamma_{212,1} = \frac{1}{2}\left[\frac{f'}{R} + g\right](X_1^2 + X_2^2) + f + \frac{g'}{R}X_1^2X_2^2, \\
\Gamma_{112,3} &= \Gamma_{312,1} = \frac{1}{2}\left[\frac{f'}{R} + g\right]X_2X_3 + \frac{g'}{R}X_1^2X_2X_3, \\
\Gamma_{212,3} &= \Gamma_{312,2} = \frac{1}{2}\left[\frac{f'}{R} + g\right]X_1X_3 + \frac{g'}{R}X_1X_2^2X_3,
\end{aligned} \tag{34}$$

where f and g are functions of $R \doteq |\mathbf{X}|$ given by

$$f = \begin{cases} A_1 + A_3R^2 & \text{if} \quad 0 \le R \le R_{\mathrm{p}} \\ B_1 + \dfrac{B_2}{R^5} + B_3R^2 + \dfrac{B_4}{R^3} & \text{if} \quad R_{\mathrm{p}} \le R \le 1 \end{cases} \tag{35}$$

and

$$g = \begin{cases} -\dfrac{2(6\kappa_{\mathrm{p}} + 17\mu_{\mathrm{p}})}{15\kappa_{\mathrm{p}} + 11\mu_{\mathrm{p}}}A_3 & \text{if} \quad 0 \le R \le R_{\mathrm{p}} \\ -5\dfrac{B_2}{R^7} - \dfrac{2(6\kappa + 17\mu)}{15\kappa + 11\mu}B_3 & \\ \quad + \left[1 + \dfrac{3\kappa}{\mu}\right]\dfrac{B_4}{R^5} & \text{if} \quad R_{\mathrm{p}} \le R \le 1. \end{cases} \tag{36}$$

In these expressions, use has been made of the notation $f'(R) \doteq \mathrm{d}f(R)/\mathrm{d}R$, $g'(R) \doteq \mathrm{d}g(R)/\mathrm{d}R$, and A_1, A_3, B_1, B_2, B_3, B_4 are constants that depend on the shear and bulk moduli of the particles, μ_{p}, κ_{p}, the matrix, μ, κ, the suspension, $\tilde{\mu}$, $\tilde{\kappa}$, as well as on the concentration of particles, $c \doteq R_{\mathrm{p}}^3$. Because of their bulkiness, the explicit form of these constants is deferred to Appendix A.

Next, we present the solution for the combination of components $\Gamma_{ikk,j}$. This combination is needed in the computation of the effective bulk modulus (30) and the effective electrostrictive coefficient (33). Physically, it characterizes the elastic response of the suspension under hydrostatic loading. It can be compactly written as follows:

$$\Gamma_{ikk,j} = \left[d + \frac{e}{R^3}\right]\delta_{ij} - \frac{3e}{R^5}X_iX_j, \tag{37}$$

where

$$d = \begin{cases} d_{\mathrm{p}} & \text{if} \quad 0 \le R \le R_{\mathrm{p}} \\ d_{\mathrm{m}} & \text{if} \quad R_{\mathrm{p}} \le R \le 1 \end{cases} \tag{38}$$

with

$$d_{\mathrm{p}} = \frac{(3\kappa+4\mu)(3\tilde{\kappa}+4\tilde{\mu})}{12c(\kappa_{\mathrm{p}}-\kappa)(\mu-\tilde{\mu})+(3\kappa+4\tilde{\mu})(3\kappa_{\mathrm{p}}+4\mu)}, \tag{39}$$

$$d_{\mathrm{m}} = \frac{(3\kappa_{\mathrm{p}}+4\mu)(3\tilde{\kappa}+4\tilde{\mu})}{12c(\kappa_{\mathrm{p}}-\kappa)(\mu-\tilde{\mu})+(3\kappa+4\tilde{\mu})(3\kappa_{\mathrm{p}}+4\mu)}, \tag{40}$$

and

$$e = \begin{cases} 0 & \text{if} \quad 0 \le R \le R_{\mathrm{p}} \\ e_{\mathrm{m}} & \text{if} \quad R_{\mathrm{p}} \le R \le 1 \end{cases} \tag{41}$$

with

$$e_{\mathrm{m}} = \frac{3c(\kappa-\kappa_{\mathrm{p}})(3\tilde{\kappa}+4\tilde{\mu})}{12c(\kappa_{\mathrm{p}}-\kappa)(\mu-\tilde{\mu})+(3\kappa+4\tilde{\mu})(3\kappa_{\mathrm{p}}+4\mu)}. \tag{42}$$

B. The solution for γ

Finally, we present the solution for the gradient $\gamma_{i,j}$, which is needed in the computation of the effective permittivity (31), as well as in the computation of the two effective electrostrictive coefficients (32) and (33). Physically, it characterizes the dielectric response of the suspension under an arbitrary affine electric field. The solution is given by

$$\gamma_{i,j} = -\left[a + \frac{b}{R^3}\right]\delta_{ij} + \frac{3b}{R^5}X_iX_j, \tag{43}$$

where

$$a = \begin{cases} a_{\mathrm{p}} & \text{if} \quad 0 \le R \le R_{\mathrm{p}} \\ a_{\mathrm{m}} & \text{if} \quad R_{\mathrm{p}} \le R \le 1 \end{cases} \tag{44}$$

with

$$a_{\mathrm{p}} = \frac{-9\varepsilon\tilde{\varepsilon}}{2c(\tilde{\varepsilon}-\varepsilon)(\varepsilon-\varepsilon_{\mathrm{p}})+(2\tilde{\varepsilon}+\varepsilon)(2\varepsilon+\varepsilon_{\mathrm{p}})}, \tag{45}$$

$$a_{\mathrm{m}} = \frac{-3(2\varepsilon+\varepsilon_{\mathrm{p}})\tilde{\varepsilon}}{2c(\tilde{\varepsilon}-\varepsilon)(\varepsilon-\varepsilon_{\mathrm{p}})+(2\tilde{\varepsilon}+\varepsilon)(2\varepsilon+\varepsilon_{\mathrm{p}})}, \tag{46}$$

and

$$b = \begin{cases} 0 & \text{if} \quad 0 \le R \le R_{\mathrm{p}} \\ b_{\mathrm{m}} & \text{if} \quad R_{\mathrm{p}} \le R \le 1 \end{cases} \tag{47}$$

with

$$b_{\mathrm{m}} = \frac{3c(\varepsilon_{\mathrm{p}}-\varepsilon)\tilde{\varepsilon}}{2c(\tilde{\varepsilon}-\varepsilon)(\varepsilon-\varepsilon_{\mathrm{p}})+(2\tilde{\varepsilon}+\varepsilon)(2\varepsilon+\varepsilon_{\mathrm{p}})}. \tag{48}$$

C. The effective electromechanical constants

We are now in a position to determine the effective electromechanical constants $\tilde{\mu}$, $\tilde{\kappa}$, $\tilde{\varepsilon}$, $\tilde{m}_K$, $\tilde{m}_J$ in (16) that characterize the overall elastic dielectric response of the suspension. Making use of the results (34), (37), (43) for the local fields $\Gamma_{i12,j}$, $\Gamma_{ikk,j}$, $\gamma_{i,j}$, it is a simple matter to carry out the integrals (29) through (33). Since the fields $\Gamma_{i12,j}$, $\Gamma_{ikk,j}$, $\gamma_{i,j}$ depend on $\tilde{\mu}$, $\tilde{\kappa}$, $\tilde{\varepsilon}$, expressions (29), (30), and (31) render polynomial equations for such effective constants. Only one root of these equations turns out to be physical. On the other hand, expressions (32) and (33) directly render explicit solutions for $\tilde{m}_K$ and $\tilde{m}_J$. After some algebraic manipulation, the result for all five effective electromechanical constants can be written as

$$\begin{aligned}
\tilde{\mu} &= \frac{q_2+\sqrt{q_2^2+4q_1q_3}}{2q_1}\mu, \quad \tilde{\kappa} = \kappa + \frac{c(3\kappa+4\mu)(\kappa_{\mathrm{p}}-\kappa)}{3\kappa_{\mathrm{p}}+4\mu-3c(\kappa_{\mathrm{p}}-\kappa)}, \quad \tilde{\varepsilon} = \varepsilon + \frac{3c(\varepsilon_{\mathrm{p}}-\varepsilon)\varepsilon}{(2+c)\varepsilon+(1-c)\varepsilon_{\mathrm{p}}}, \\
\tilde{m}_K &= 2A_1a_{\mathrm{p}}^2c\,m_{K_{\mathrm{p}}} + \frac{42A_3a_{\mathrm{p}}^2c^{5/3}(3\kappa_p+\mu_{\mathrm{p}})m_{K_{\mathrm{p}}}}{75\kappa_p+55\mu_{\mathrm{p}}} + \frac{2}{5}B_1\left(\frac{1}{c}-1\right)(5a_{\mathrm{m}}^2c+b_{\mathrm{m}}^2)m_K - \frac{27}{5}B_2b_{\mathrm{m}}^2\left(1-\frac{1}{c^{8/3}}\right)m_K \\
&\quad + \frac{6B_3\left[7a_{\mathrm{m}}^2(c^{1/3}-c^2)(3\kappa+\mu)+7a_{\mathrm{m}}b_{\mathrm{m}}(c-c^{1/3})(3\kappa-2\mu)+b_{\mathrm{m}}^2(1-c^{1/3})(15\kappa+11\mu)\right]m_K}{5c^{1/3}(15\kappa+11\mu)} \\
&\quad - \frac{252B_3b_{\mathrm{m}}\mu\left[a_{\mathrm{m}}c+b_{\mathrm{m}}-c^{1/3}(a_{\mathrm{m}}+b_{\mathrm{m}})\right]m_J}{5c^{1/3}(15\kappa+11\mu)} - \frac{B_4b_{\mathrm{m}}(1-c)\left[b_{\mathrm{m}}(1+c)(15\kappa+4\mu)-2a_{\mathrm{m}}c(3\kappa+8\mu)\right]m_K}{5c^2\mu} \\
&\quad - \frac{2B_4b_{\mathrm{m}}(1-c)(b_{\mathrm{m}}c-4a_{\mathrm{m}}c+b_{\mathrm{m}})m_J}{5c^2}, \\
\tilde{m}_J &= (1-c)(\tilde{\varepsilon}-\varepsilon)^2(\tilde{\kappa}-\kappa)\left[\frac{(3\kappa_{\mathrm{p}}+4\mu)\left[2(2+c)\varepsilon^2+4(1-c)\varepsilon\varepsilon_{\mathrm{p}}+(1+2c)\varepsilon_{\mathrm{p}}^2\right]m_J}{9c^3\varepsilon^2(\varepsilon_{\mathrm{p}}-\varepsilon)^2(\kappa_{\mathrm{p}}-\kappa)(3\kappa+4\mu)} + \frac{\left[(7-c)\varepsilon+(5+c)\varepsilon_{\mathrm{p}}\right]m_K}{3c^2\varepsilon^2(\varepsilon_{\mathrm{p}}-\varepsilon)(3\kappa+4\mu)}\right] \\
&\quad + \frac{(\tilde{\varepsilon}-\varepsilon)^2(\tilde{\kappa}-\kappa)m_{J_{\mathrm{p}}}}{c^2(\varepsilon_{\mathrm{p}}-\varepsilon)^2(\kappa_{\mathrm{p}}-\kappa)},
\end{aligned} \tag{49}$$

where q_1, q_2, q_3 are given by expressions (B1) in Appendix B, and it is recalled that A_1, A_3, B_1, B_2, B_3, B_4 are defined in Appendix A, while a_{p}, a_{m}, b_{m} are given by expressions (45), (46), and (48). A number of theoretical and practical remarks are of note:

1. The solution (49) is valid for any choice of the shear moduli μ, μ_{p}, bulk moduli κ, κ_{p}, permittivities ε, ε_{p}, and electrostrictive coefficients m_K, $m_{K_{\mathrm{p}}}$, m_J, $m_{J_{\mathrm{p}}}$ characterizing the elastic dielectric response of the underlying rubber matrix and the particles, as well as for any choice of the concentration of particles $c \in [0, 1]$.
2. There are several limiting cases contained in (49) worth exploring in detail. In this paper, we restrict ourselves to recording explicitly two of them. In the fundamental limit when the particles are present in dilute concentration as $c \to 0+$, the effective constants (49) reduce asymptotically to

$$
\begin{aligned}
\tilde{\mu}^{\,\mathrm{dil}} &= \mu + \frac{5(3\kappa+4\mu)(\mu_{\mathrm{p}}-\mu)\mu}{(9\kappa+8\mu)\mu+6(\kappa+2\mu)\mu_{\mathrm{p}}}\,c, \quad \tilde{\kappa}^{\,\mathrm{dil}} = \kappa + \frac{(3\kappa+4\mu)(\kappa_{\mathrm{p}}-\kappa)}{3\kappa_{\mathrm{p}}+4\mu}\,c, \quad \tilde{\varepsilon}^{\,\mathrm{dil}} = \varepsilon + \frac{3(\varepsilon_{\mathrm{p}}-\varepsilon)\varepsilon}{2\varepsilon+\varepsilon_{\mathrm{p}}}\,c,\\
\tilde{m}_K^{\,\mathrm{dil}} &= m_K + \left[\frac{45\varepsilon^2\mu(3\kappa+4\mu)m_{K_{\mathrm{p}}}}{(2\varepsilon+\varepsilon_{\mathrm{p}})^2\left[6\mu_{\mathrm{p}}(\kappa+2\mu)+\mu(9\kappa+8\mu)\right]} + \frac{2\mu(\varepsilon_{\mathrm{p}}-\varepsilon)(7\varepsilon+5\varepsilon_{\mathrm{p}})(\mu_{\mathrm{p}}-\mu)m_J}{(2\varepsilon+\varepsilon_{\mathrm{p}})^2\left[6\mu_{\mathrm{p}}(\kappa+2\mu)+\mu(9\kappa+8\mu)\right])}\right.\\
&\quad \left. - \frac{\mu\left[\varepsilon^2(81\kappa+77\mu)+\varepsilon\varepsilon_{\mathrm{p}}(54\kappa+86\mu)+17\varepsilon_{\mathrm{p}}^2\mu\right]-\mu_{\mathrm{p}}(\varepsilon_{\mathrm{p}}-\varepsilon)[36\varepsilon\kappa+79\varepsilon\mu+18\varepsilon_{\mathrm{p}}\kappa+41\varepsilon_{\mathrm{p}}\mu]}{(2\varepsilon+\varepsilon_{\mathrm{p}})^2\left[6\mu_{\mathrm{p}}(\kappa+2\mu)+\mu(9\kappa+8\mu)\right]}m_K\right]c,\\
\tilde{m}_J^{\,\mathrm{dil}} &= m_J + \left[\frac{9\varepsilon^2(3\kappa+4\mu)m_{J_{\mathrm{p}}}}{(2\varepsilon+\varepsilon_{\mathrm{p}})^2(3\kappa_{\mathrm{p}}+4\mu)} + 3\left(\frac{\kappa_{\mathrm{p}}-\kappa}{3\kappa_{\mathrm{p}}+4\mu} - \frac{2\varepsilon^2+2\varepsilon\varepsilon_{\mathrm{p}}-\varepsilon_{\mathrm{p}}^2}{(2\varepsilon+\varepsilon_{\mathrm{p}})^2}\right)m_J + \frac{3(\varepsilon_{\mathrm{p}}-\varepsilon)(7\varepsilon+5\varepsilon_{\mathrm{p}})(\kappa_{\mathrm{p}}-\kappa)m_K}{(2\varepsilon+\varepsilon_{\mathrm{p}})^2(3\kappa_{\mathrm{p}}+4\mu)}\right]c
\end{aligned}
\tag{50}
$$

to $O(c)$. These results constitute a generalization of the classical results of Eshelby[27] and Maxwell[28] for the purely elastic and purely dielectric overall response of a dilute suspension of spherical particles to the coupled and nonlinear realm of elastic dielectric properties.

3. When synthesized under typical conditions, rubber can be regarded to be incompressible,[29] so that $\kappa = +\infty$, and not to exhibit "material" electrostriction,[30] so that $m_K = m_J = \varepsilon$. When compared with rubber, furthermore, typical filler particles such as ceramics and metals can be regarded to be mechanically rigid and also not to exhibit "material" electrostriction, so that $\mu_{\mathrm{p}}, \kappa_{\mathrm{p}} = +\infty$ and $m_{K_{\mathrm{p}}} = m_{J_{\mathrm{p}}} = \varepsilon_{\mathrm{p}}$. For this practically relevant choice of rubber and particle properties, the effective constants (49) reduce to the more compact form

$$
\begin{aligned}
\tilde{\mu}^{\,\mathrm{rig,inc}} &= \mu + \frac{35c\mu}{7-15c+8c^{10/3}+\sqrt{49+c[14+c(8c^{2/3}(8c^4-2c^{5/3}-161c^{2/3}+294)-1175)]}}, \quad \tilde{\kappa}^{\,\mathrm{rig,inc}} = +\infty,\\
\tilde{\varepsilon}^{\,\mathrm{rig,inc}} &= \varepsilon + \frac{3c(\varepsilon_{\mathrm{p}}-\varepsilon)\varepsilon}{(2+c)\varepsilon+(1-c)\varepsilon_{\mathrm{p}}},\\
\tilde{m}_K^{\,\mathrm{rig,inc}} &= \varepsilon + (\tilde{\varepsilon}^{\,\mathrm{rig,inc}}-\varepsilon)^2\left[\frac{\mu}{8(1-c^{2/3})\varepsilon(\tilde{\mu}^{\,\mathrm{rig,inc}}-\mu)} + \frac{(1+c^{1/3})(17-8c)-3c^{2/3}}{60(1+c^{1/3})c\,\varepsilon} + \frac{1}{c(\varepsilon_{\mathrm{p}}-\varepsilon)}\right],\\
\tilde{m}_J^{\,\mathrm{rig,inc}} &= \varepsilon + \frac{9c\left(\varepsilon_{\mathrm{p}}^2-\varepsilon^2\right)\varepsilon}{\left[(2+c)\varepsilon+(1-c)\varepsilon_{\mathrm{p}}\right]^2},
\end{aligned}
\tag{51}
$$

where use has been made of the inequality $\kappa_{\mathrm{p}} \gg \kappa$, typical of standard ceramic and metallic filler particles when compared to rubber, in the computation of the electrostrictive coefficient $(51)_5$.

4. As expected, the effective shear modulus $(49)_1$ agrees identically with the result originally derived by Christensen and Lo,[31] and later proved to be realizable by Avellaneda,[23] for a differential coated sphere assemblage. Further, the effective bulk modulus $(49)_2$ and effective permittivity $(49)_3$ agree identically with the classical results of Hashin[22] and Hashin and Shtrikman[32] for arbitrary (not necessarily differential) coated sphere assemblages. As a corollary, the results $(49)_2$ and $(49)_3$ also agree with one of the Hashin-Shtrikman bounds for the bulk

modulus and permittivity of two-phase composites with isotropic (not necessarily particulate) microstructures when the elastic and dielectric properties of the matrix and particles are well ordered.

5. Similar to the results $(49)_2$ and $(49)_3$, the result $(49)_5$ for the effective electrostrictive coefficient $\tilde{m}_J$ can be shown (via a neutral inclusion argument) to be exact not just for a differential coated sphere assemblage but for any coated sphere assemblage. This is not true for the result $(49)_4$ for the electrostrictive coefficient $\tilde{m}_K$, which, similar to the effective shear modulus $(49)_1$, is exact only for a differential coated sphere assemblage.
6. While exact for a suspension with spherical particles of infinitely many sizes, the results $(49)_1$, $(49)_2$, and $(49)_3$ have been shown[33–35] to accurately describe as well the elastic and the dielectric response of isotropic suspensions with spherical particles of the same size up to relatively large particle concentrations sufficiently away from percolation (at least up to about $c = 0.25$, but possibly up to larger values of c depending on the heterogeneity contrast). By means of sample comparisons with finite-element simulations, in the next section we show that the results $(49)_4$ and $(49)_5$ for the electrostrictive coefficients $\tilde{m}_K$ and $\tilde{m}_J$ are also accurately descriptive of isotropic suspensions with spherical particles of the same size sufficiently away from percolation (at least up to about $c = 0.2$).

IV. SAMPLE RESULTS AND DISCUSSION

In the sequel, we present sample results based on the analytical solution (49) and confront them to finite-element simulations for the overall elastic dielectric response of rubber filled with a random isotropic distribution of monodisperse spherical particles; the details of the simulations are deferred for presentation elsewhere.[35] A first aim of these sample results is to demonstrate the applicability of the solution (49) to isotropic suspensions with spherical particles of the same size over a large range of concentrations of particles. A second aim is to deploy the solution (49) to gain insight into experimental findings. In particular, we seek to scrutinize the drastic enhancement in electrostrictive properties observed experimentally in elastomers filled with high-permittivity and (semi-)conducting particles,[3–9] in spite of the fact that such particles were mechanically much stiffer than the dielectric elastomers to which they were added.

All of the results that follow pertain to suspensions wherein the rubber is incompressible, $\kappa = +\infty$, and the particles are mechanically rigid, $\mu_p, \kappa_p = +\infty$, and of infinite permittivity with $\varepsilon_p = m_{K_p} = m_{J_p} = +\infty$. The rationale behind this choice of material parameters is that they are representative of many of the dielectric elastomer composites studied experimentally, such as for instance those of Huang *et al.*[5] where the elastomer is polyurathane ($\mu = 9$ MPa, $\kappa = 5$ GPa, $\varepsilon = 8\varepsilon_0$) and the particles are made out of a metallophthalocyanine oligomer ($\mu_p = 1$ GPa, $\kappa_p = 100$ GPa, $\varepsilon_p = 10^4\varepsilon_0$), and those of Liu *et al.*,[9] where the elastomer is silicone rubber ($\mu = 0.22$ MPa, $\kappa = 1$ GPa, $\varepsilon = 3.2\varepsilon_0$) and the particles are made out of titania ($\mu_p = 110$ GPa, $\kappa_p = 220$ GPa, $\varepsilon_p = 114\varepsilon_0$).

Figure 1 shows results for the effective shear modulus $\tilde{\mu}$, normalized by the shear modulus of the rubber μ, the effective permittivity $\tilde{\varepsilon}$, normalized by the permittivity of the rubber ε, and the effective electrostrictive coefficients $\tilde{m}_K/\varepsilon$, $\tilde{m}_J/\varepsilon$, normalized by ε, all as functions of the concentration of particles c, as predicted by the analytical solution (49) and by the finite-element simulations. It is plain from

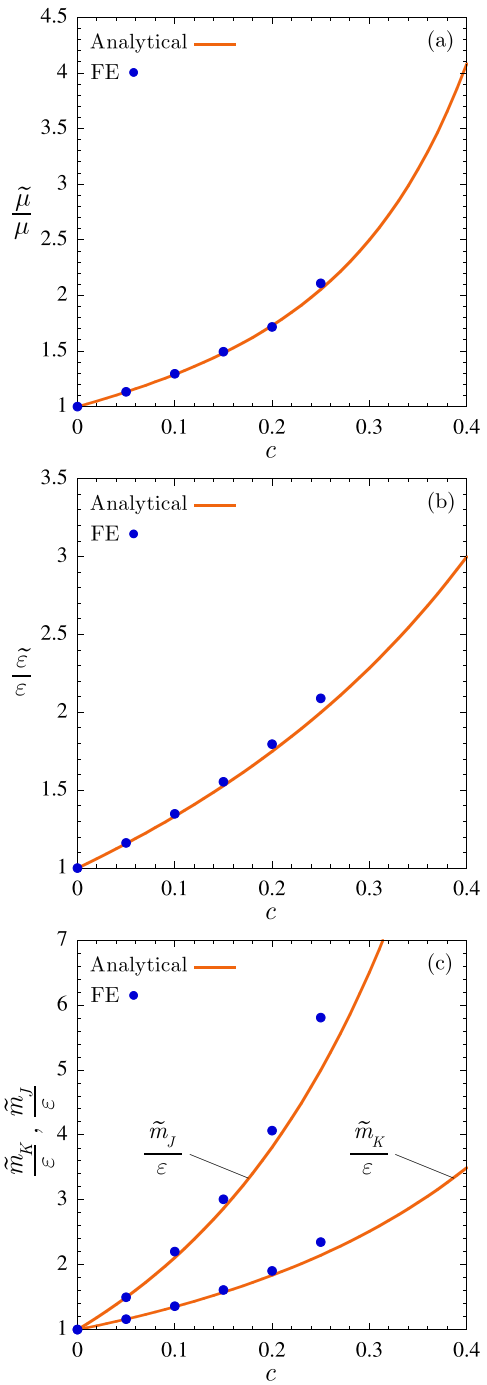

FIG. 1. (a) Effective shear modulus $\tilde{\mu}/\mu$, (b) permittivity $\tilde{\varepsilon}/\varepsilon$, and (c) electrostrictive coefficients $\tilde{m}_K/\varepsilon$, $\tilde{m}_J/\varepsilon$ of an isotropic suspension of rigid ($\mu_p, \kappa_p = +\infty$), infinite-permittivity ($\varepsilon_p = +\infty$), spherical particles embedded in an incompressible rubber ($\kappa = +\infty$) with shear modulus μ and permittivity ε, all as functions of the concentration of particles c. The solid line corresponds to the analytical solution (49), whereas the solid circles correspond to finite-element simulations for a random isotropic suspension of monodisperse spherical particles.

the figure that the analytical solution (solid line), even though exact for a suspension with particles of infinitely many sizes, serves also to describe the elastic dielectric response of suspensions with particles of the same size (solid circles) up to relatively large particle concentrations of about $c=0.2$. In this comparison, it is of note that the effective shear modulus $\tilde{\mu}$ and the effective permittivity $\tilde{\varepsilon}$ are in accord for both (poly- and mono-disperse) microstructures up to about $c=0.25$, and that the effective electrostrictive coefficients $\tilde{m}_K$ and $\tilde{m}_J$ start showing sizable differences between the two microstructures at smaller particle concentrations of about $c=0.2$.

As expected on physical grounds, Figs. 1(a) and 1(b) show that the addition of rigid infinite-permittivity particles enhances both the shear stiffness and permittivity over those of the pure (unfilled) rubber. Figure 1(c) confirms that this is also the case for the electrostrictive coefficients $\tilde{m}_K$ and $\tilde{m}_J$. It is apparent from Eq. (19) that these trends set up a direct competition of effects for the overall electrostriction capabilities of the suspension. To see which enhancement proves dominant, if the enhancement in stiffness (which makes the material less deformable) or that in permittivity and electrostrictive coefficients (which makes the material more prone to deform under the application of an electric field), we consider, as in a typical experiment, a uniaxial electric field of magnitude $\bar{E}$ applied in the $\mathbf{e}_3$ direction say. In the present context of incompressible rubber and rigid particles with infinite permittivity, the electrostriction (23) in the direction of the applied field specializes then to

$$\bar{H}_{33}=-\left[\frac{\varepsilon}{3\tilde{\mu}^{\,\mathrm{rig,inc}}}-\frac{c[8c^{4/3}+3c^{2/3}+8c-17(1+c^{1/3})]\dfrac{\varepsilon}{\tilde{\mu}^{\,\mathrm{rig,inc}}}}{20(1+c^{1/3})(1-c)^2\left[1-\dfrac{\mu}{\tilde{\mu}^{\,\mathrm{rig,inc}}}\right]}-\frac{c\left(\dfrac{17}{20}+\dfrac{2}{5}c^{5/3}-c^{2/3}-\dfrac{5}{8}c\right)\dfrac{\varepsilon}{\tilde{\mu}^{\,\mathrm{rig,inc}}}}{(1-c^{8/3}+2c^{5/3}-c^{2/3}+c^2-2c)\left[\dfrac{\tilde{\mu}^{\,\mathrm{rig,inc}}}{\mu}-1\right]}\right]\bar{E}^2, \tag{52}$$

where it is recalled that the effective shear modulus $\tilde{\mu}^{\,\mathrm{rig,inc}}$ is given by expression $(51)_1$; because of the overall incompressibility of the suspension, the electrostriction (22) transverse to the applied field is simply given by $\bar{H}_{11}=\bar{H}_{22}=-\bar{H}_{33}/2$. In the absence of particles when $c=0$, expression (52) reduces to the electrostriction of the pure rubber

$$H^m_{33}=-\frac{\varepsilon}{3\mu}\bar{E}^2, \tag{53}$$

and so the ratio

$$\frac{\bar{H}_{33}}{H^m_{33}}=\frac{\mu}{\tilde{\mu}^{\,\mathrm{rig,inc}}}-\frac{3c[8c^{4/3}+3c^{2/3}+8c-17(1+c^{1/3})]\dfrac{\mu}{\tilde{\mu}^{\,\mathrm{rig,inc}}}}{20(1+c^{1/3})(1-c)^2\left[1-\dfrac{\mu}{\tilde{\mu}^{\,\mathrm{rig,inc}}}\right]}-\frac{3c\left(\dfrac{17}{20}+\dfrac{2}{5}c^{5/3}-c^{2/3}-\dfrac{5}{8}c\right)\dfrac{\mu}{\tilde{\mu}^{\,\mathrm{rig,inc}}}}{(1-c^{8/3}+2c^{5/3}-c^{2/3}+c^2-2c)\left[\dfrac{\tilde{\mu}^{\,\mathrm{rig,inc}}}{\mu}-1\right]}, \tag{54}$$

provides a direct measure of the effect that the addition of particles has on the electrostrictive properties. Figure 2 shows a plot of this ratio as a function of the concentration of particles c. Akin to Fig. 1, the solid line corresponds to the analytical solution (54), whereas the solid circles correspond to finite-element simulations for a random isotropic suspension of monodisperse spherical particles. Again, both results are seen to be in good agreement up to particle concentrations of about $c=0.2$.

An immediate observation from Fig. 2 is that the addition of rigid, infinite-permittivity, spherical particles monotonically enhances the electrostrictive capabilities of rubber ($\bar{H}_{33}/H^m_{33}>1$) up to particle concentrations of about $c=0.18$. The further addition of particles leads to a lesser enhancement, and beyond about $c=0.3$, to a decrease in electrostriction ($\bar{H}_{33}/H^m_{33}<1$) when compared to the pure rubber. These results reveal that the enhancement in permittivity and electrostrictive coefficients granted by the addition of spherical particles dominates over the enhancement in

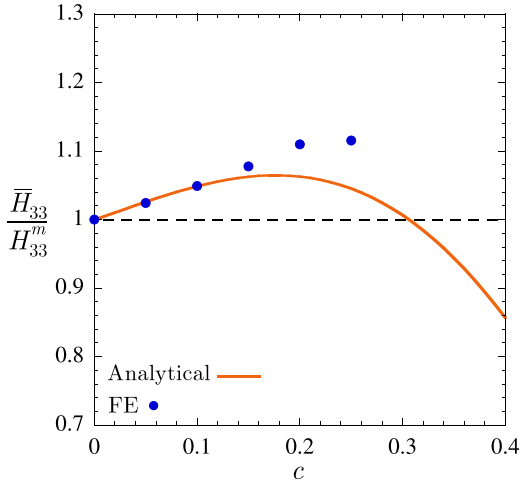

FIG. 2. The electrostriction $\bar{H}_{33}/H^m_{33}$ of an isotropic suspension of rigid ($\mu_\mathrm{p}, \kappa_\mathrm{p}=+\infty$), infinite-permittivity ($\varepsilon_\mathrm{p}=+\infty$), spherical particles embedded in an incompressible rubber ($\kappa=+\infty$), as a function of the concentration of particles c. The solid line corresponds to the analytical solution (54), whereas the solid circles correspond to finite-element simulations for a random isotropic suspension of monodisperse spherical particles.

stiffness, provided that the concentration of particles is below certain threshold, here $c = 0.3063$. As the concentration exceeds this threshold, the enhancement in stiffness becomes dominant and the electrostriction capabilities of the suspension are outperformed by those of the pure rubber.

Now, in quantitative terms, the enhancement granted by the addition of particles is admittedly modest. Indeed, the maximum value of the ratio of electrostrictions, occurring at $c = 0.1764$, is given by $\bar{H}_{33}/H^m_{33} = 1.065$. This is in disagreement with most experimental findings [3–9] which have reported enhancements in electrostriction ranging from several tens to several thousands of a percent for small particle concentrations $c < 0.1$, far exceeding the maximum enhancement of 6.5% found here.

A plausible explanation for at least part of the difference between the theoretical solution constructed here and the experiments is that the microstructures in the experiments might contain anisotropic particle aggregates and thus are not well approximated by a random isotropic distribution of spherical particles. Indeed, Tian[36] and Tian *et al.*[12] have shown via some 2D examples that anisotropic particles, when distributed appropriately, may lead to larger spatial fluctuations of the electric field and hence may enhance the effective electrostrictive response of two-phase particulate composites in a more substantial way than circular particles.

It seems unlikely, however, that the drastic enhancement observed in the experiments may be solely due to the presence of anisotropic particle aggregates. We conjecture, alternatively, that this extreme behavior may be due to the presence of high-permittivity interphases and/or interphasial free charges around the filler particles. Indeed, it is well known that in filled elastomers, the "anchoring" of the polymeric chains of the matrix onto the filler particles forces the chains into conformations that are very different from those in the bulk, hence resulting in "interphases" with very different mechanical and physical properties than the elastomer in the bulk.[13,37] Furthermore, free charges in such interphases may be present from the outset because of the fabrication process of the material. They may also be injected from the particles upon the application of an electric field (note that in the experiments of Huang *et al.*,[5] where an enhancement in electrostriction of 2000% was reported, the particles were made out of an organic semi-conductor coated by an anionic polyacrylic acid). Whatever their origin, the presence of interphasial charges has been recently shown to have the potential to lead to extreme enhancements of the overall dielectric properties of particulate composites and, by the same token, extreme enhancements of their electrostrictive properties.[14]

ACKNOWLEDGMENTS

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APPENDIX A: 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 (35) and (36) are defined by the continuity of the fields Γ_{rkl} and $L_{ijrs}\Gamma_{rkl,s}X_j$ across the two material interfaces at $R = R_p$, 1, and by the boundary condition at infinity where $\Gamma_{ikl} = \delta_{ik}\delta_{jl}X_j$. These conditions lead to six linear equations for the six constants, namely,

$$
\begin{aligned}
\mathcal{E}_1 &= -A_1 - c^{2/3}A_3 + B_1 + \frac{B_2}{c^{5/3}} + c^{2/3}B_3 + \frac{B_4}{c} = 0,\\
\mathcal{E}_2 &= \frac{2\left(6\kappa_p + 17\mu_p\right)A_3}{15\kappa_p + 11\mu_p} - \frac{5B_2}{c^{7/3}} - \frac{2(6\kappa + 17\mu)B_3}{15\kappa + 11\mu} + \frac{(3\kappa + \mu)B_4}{c^{5/3}\mu} = 0,\\
\mathcal{E}_3 &= -\mu_p A_1 - \frac{63c^{2/3}\mu_p^2 A_3}{19\left(15\kappa_p + 11\mu_p\right)} + \mu B_1 + \frac{84\mu B_2}{19c^{5/3}} + \frac{63c^{2/3}\mu^2 B_3}{19(15\kappa + 11\mu)} - \frac{(135\kappa + 64\mu)B_4}{38c} = 0,\\
\mathcal{E}_4 &= \frac{\mu_p\left(57\kappa_p + 4\mu_p\right)A_3}{15\kappa_p + 11\mu_p} + \frac{20\mu B_2}{c^{7/3}} - \frac{\mu(57\kappa + 4\mu)B_3}{15\kappa + 11\mu} - \frac{4(3\kappa + \mu)B_4}{c^{5/3}} = 0,\\
\mathcal{E}_5 &= \left[\frac{6\mu(\tilde{\kappa} + 2\tilde{\mu})}{\tilde{\mu}(9\tilde{\kappa} + 8\tilde{\mu})} + 1\right]B_1 + \left(1 - \frac{\mu}{\tilde{\mu}}\right)B_2 + \left[\frac{28\mu(3\kappa + \mu)}{(15\kappa + 11\mu)(9\tilde{\kappa} + 8\tilde{\mu})} + \frac{3\mu(15\kappa + 4\mu)}{60\kappa\tilde{\mu} + 44\mu\tilde{\mu}} + 1\right]B_3\\
&\quad + \left[-\frac{2(9\kappa + 8\mu)}{3(9\tilde{\kappa} + 8\tilde{\mu})} - \frac{\mu}{3\tilde{\mu}} + 1\right]B_4 - \frac{5(3\tilde{\kappa} + 4\tilde{\mu})}{2(9\tilde{\kappa} + 8\tilde{\mu})} = 0,\\
\mathcal{E}_6 &= 5\left(\frac{\mu}{\tilde{\mu}} - 1\right)B_2 - \frac{\left[\kappa(57\mu + 48\tilde{\mu}) + 4\mu(\mu + 34\tilde{\mu})\right]B_3}{60\kappa\tilde{\mu} + 44\mu\tilde{\mu}} - \frac{(3\kappa + \mu)(\mu - \tilde{\mu})B_4}{\mu\tilde{\mu}} = 0.
\end{aligned}
\tag{A1}
$$

While it is simple matter to compute the unique solution to this system of linear equations for A_1, A_3, B_1, B_2, B_3, B_4, we do not report it here because of the bulkiness of the final expressions.

APPENDIX B: EXPRESSIONS FOR q_1, q_2, q_3

The expressions for q_1, q_2, q_3 in the result $(49)_1$ for the effective shear modulus $\tilde{\mu}$ are given by

$$q_1 = 6c^{10/3}\eta_1\left(2+\frac{\kappa}{\mu}\right) - 126c^{5/3}\eta_3\left(1+\frac{3\kappa}{\mu}\right)^2 + c^{7/3}\eta_2 + 225c\,\eta_3\left(\frac{3\kappa^2}{\mu^2}+\frac{4\kappa}{\mu}+3\right) + \frac{3\eta_3\left(\frac{6\kappa}{\mu}+17\right)\left[6\frac{\mu_p}{\mu}\left(\frac{\kappa}{\mu}+2\right)+\frac{9\kappa}{\mu}+8\right]}{1-\frac{\mu_p}{\mu}},$$

$$q_2 = c^{10/3}\eta_1\left(4-\frac{3\kappa}{\mu}\right) - 252c^{5/3}\eta_3\left(\frac{3\kappa}{\mu}+1\right)^2 + 2c^{7/3}\eta_2 + \frac{75}{4}c\,\eta_3\left(\frac{3\kappa}{\mu}-2\right)\left(\frac{15\kappa}{\mu}+8\right) - \frac{9\eta_3\left(\frac{3\kappa}{\mu}-44\right)\left[\frac{6\mu_p}{\mu}\left(\frac{\kappa}{\mu}+2\right)+\frac{9\kappa}{\mu}+8\right]}{8\left(1-\frac{\mu_p}{\mu}\right)},$$

$$q_3 = c^{10/3}\eta_1\left(8+\frac{9\kappa}{\mu}\right) + 126c^{5/3}\eta_3\left(\frac{3\kappa}{\mu}+1\right)^2 - c^{7/3}\eta_2 - \frac{75}{8}c\,\eta_3\left(\frac{81\kappa^2}{\mu^2}+\frac{60\kappa}{\mu}+8\right) + \frac{3\eta_3\left(\frac{57\kappa}{\mu}+4\right)\left[\frac{6\mu_p}{\mu}\left(\frac{\kappa}{\mu}+2\right)+\frac{9\kappa}{\mu}+8\right]}{8\left(1-\frac{\mu_p}{\mu}\right)}, \tag{B1}$$

where

$$\eta_1 = 3\left(1-\frac{\mu_p}{\mu}\right)\left[\frac{3\kappa\left(\frac{114\kappa_p\left(1-\frac{\mu_p}{\mu}\right)}{\mu}+\frac{\mu_p\left(323-\frac{8\mu_p}{\mu}\right)}{\mu}\right)}{\mu} + \frac{3\kappa_p\left(8-\frac{323\mu_p}{\mu}\right)}{\mu} + \frac{68\mu_p\left(1-\frac{\mu_p}{\mu}\right)}{\mu}\right],$$

$$\eta_2 = 225\left[\frac{3\kappa^2\left[\frac{54\kappa_p}{\mu}-\frac{\mu_p^2\left(\frac{57\kappa_p}{\mu}+86\right)}{\mu^2}+\frac{3\mu_p\left(\frac{\kappa_p}{\mu}+51\right)}{\mu}-\frac{4\mu_p^3}{\mu^3}\right]}{\mu^2} + \frac{\kappa\left[\frac{120\kappa_p}{\mu}-\frac{12\mu_p^2\left(\frac{19\kappa_p}{\mu}+6\right)}{\mu^2}+\frac{\mu_p\left(340-\frac{81\kappa_p}{\mu}\right)}{\mu}-\frac{16\mu_p^3}{\mu^3}\right]}{\mu} + \frac{\kappa_p\left(16-\frac{171\mu_p^2}{\mu^2}-\frac{97\mu_p}{\mu}\right)}{\mu} + \frac{4\mu_p\left(1-\frac{\mu_p}{\mu}\right)\left(\frac{9\mu_p}{\mu}+34\right)}{3\mu}\right],$$

$$\eta_3 = \left(1-\frac{\mu_p}{\mu}\right)\left[\frac{\kappa_p\left(\frac{57\mu_p}{\mu}+48\right)}{\mu}+\frac{4\mu_p\left(\frac{\mu_p}{\mu}+34\right)}{\mu}\right]. \tag{B2}$$

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