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Liquid inclusions in soft materials: Capillary effect, mechanical stiffening and enhanced electromechanical response

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ABSTRACT

Recent experiments and modeling appear to suggest that "small" liquid inclusions may significantly stiffen soft solids. Surface energy or capillary effects are considered responsible for this rather non-intuitive phenomenology. In this work, we develop a theoretical framework and construct a simple homogenization model that unambiguously explains this phenomenon. Our theoretical construct of capillary effects is specifically intended for soft matter and differs in the manner in which the classical Gurtin-Murdoch surface elasticity theory has been widely interpreted and used in the literature. We use the developed theory to elucidate several subtle aspects pertaining to the capillary phenomenon that may not be often appreciated; specifically in regards to the developments across different scientific communities. One such key aspect is the often missed connection between the concept of surface tension as understood in the fluids and materials science community and superficial elasticity of solid surfaces. While the purely mechanical effects underpinning liquid inclusions in soft solids are of significant interest in of themselves, we hypothesize that the interplay of capillary effect, electrostatics, soft solids and *ionic* liquid inclusions may offer prospects for designing soft materials that display an unusually large size-dependent electromechanical coupling. To emphasize this, we extend the capillary framework for soft solids to electrostatics and find that electrostrictive properties may be significantly altered. Due to a rather interesting confluence of two contradictory effects, the electrostrictive deformation is only marginally altered by capillarity. However, the soft solid's maximum energy conversion capability may be enhanced by 75% or higher.

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1. Introduction

Conventional wisdom would suggest that embedding liquid inclusions in a soft elastic material (Fig. 1) ought to make the overall composite yet even softer. In addition, classical elasticity and the theories underpinning effective properties of composites (Milton, 2002) predict that the overall elastic response depends only on the shape of the embedded inclusions and not the size. These notions were decisively overturned in a recent set of intriguing experiments (Style et al., 2015a). Style

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Fig. 1. A schematic of liquid inclusions embedded in a soft solid. Recent experiments indicate that liquid inclusions may, somewhat non-intuitively, stiffen a soft solid (Style et al., 2015a). This observation is explained by invoking the effect of surface energy or capillarity and the consequent size-dependent mechanical response of the soft composite (Style et al., 2015b).

and co-workers embedded soft silicone gels of two different elastic stiffnesses with droplets (inclusions) of an ionic liquid in the size-range of a few microns. Under mechanical testing, the "stiffer" Silicone gel with an elastic modulus of 100 kPa displayed conventional behavior while the composite made with the softer Silicone gel (modulus: 1.7 kPa) exhibited overall *stiffening*. This apparent paradox was explained (Style et al., 2015b) by invoking the concept of surface energy for solids (specifically, surface tension) and the associated size-dependent mechanical response.

The premise is that a "surface"¹ has atoms that possess a coordination number, charge distribution and other attributes that are different from those of the bulk material. The solid (or for that matter, a liquid) surface may therefore be considered an independent entity from that of the underlying bulk material to which it is attached and exhibits distinct properties. Phenomenologically, this is understood from the viewpoint that the surface possesses an *excess* energy that offers its own unique mechanical signature including, in a manner of speaking, surface elasticity and a residual surface stress (or for isotropic materials, surface *tension*). Surface energy penalizes any increase of surface area (which may be due to deformation). In the case of simple fluids which are isotropic, this gives rise to the concept of a "surface tension" which, for instance, leads to the characteristic smooth spherical shape of liquid droplets.²

Classical elasticity, in absence of surface energy effects, is size-independent. Accordingly, the elastic state of an embedded inclusion—known widely as the Eshelby's inclusion problem (Eshelby, 1957,1959)—depends only upon the shape of the inclusion and consequently, as borne out by an extremely large body of literature (Li and Wang, 2008; Milton, 2002; Mura, 2013; Nemat-Nasser and Hori, 2013), the effective elastic response of a composite is also predicted to depend only on the shape and volume fraction of the inclusions and not the absolute size of the embedded objects. Surface energy effects exert

¹ We use the word "surface" somewhat broadly to also include interfaces between two materials where either of the materials may belong to solid, liquid or vapor phase.

² The concept of surface tension may be generalized to solids and is termed surface stress. As pointed out by several authors, care must be taken to not confound these two concepts c.f. Style et al. (2017). Even for isotropic solid surfaces where the surface stress may be represented by a surface-tension like scalar, due to the deformation dependent behavior of solid surface energy, there are differences in the concepts.

their influence when the surface-to-volume ratio becomes "large enough".³ Practically speaking, at least in the context of *hard* solids, this implies that except at length scales approaching a few nanometers, classical elasticity suffices (Miller and Shenoy, 2000). The experimental and pertinent theoretical work of Style and co-workers highlighted that for liquid inclusions embedded in very soft materials, the capillarity induced size-effects become manifest event for *micron* sized-inclusions and lead to the non-intuitive stiffening alluded to in the topical paragraph.

The study of surface energy and its ramifications has been independently pursued across multiple disciplines-see for example the following for representative examples-fluid mechanics (de Gennes et al., 2004), chemistry and chemical engineering (Adamson and Gast, 1967), physics (Müller and Saúl, 2004), materials (Cahn, 1989; Cammarata, 1994, 2009; Johnson, 2000; Streitz et al., 1994; Voorhees and Johnson, 2004; Weissmüller and Cahn, 1997) and of course the solid mechanics community (Dingreville et al., 2005; Duan et al., 2005a, 2005b; Fried and Todres, 2005; Gurtin and Murdoch, 1975, 1978; Henann and Anand, 2010; Henann and Bertoldi, 2014; Huang and Sun, 2007; Huang and Wang, 2006; Javili et al., 2013; Miller and Shenoy, 2000; Ou and Cherkaoui, 2007; Sharma and Ganti, 2004; Sharma et al., 2003; Sharma and Wheeler, 2007; Wang et al., 2011; Wang and Henann, 2016; Yun and Park, 2008). This broad interest is unsurprising since applications as wide-ranging as catalysis (Haiss, 2001; Müller and Saúl, 2004; Pala and Liu, 2004), self-assembly (Suo and Lu, 2000), fracture (Kim et al., 2011; Wu, 1999), wetting (de Gennes et al., 2004; Style et al., 2017), mechanical strengthening (Duan et al., 2006; Mameka et al., 2017; Sharma and Ganti, 2004; Sharma et al., 2003), phase transformation (Fischer et al., 2008). semiconductor and thin film technology (Ibach, 1997), nano-sensing (Duan et al., 2009; Mohammadi et al., 2013; Park, 2008) among many others, are influenced by surface energy related effects.⁴ On the theoretical front, the thermodynamical underpinnings of the subject date back to the famous works by Gibbs (1906) which have been frequently revisited by many researchers c.f. Cahn (1989), Cammarata (2009), Shuttleworth (1950), and Orowan (1970). Arguably the most rigorous theoretical framework for surface energy, in the spirit of classical continuum field theories, was first established by Gurtin and Murdoch (1975, 1978)⁵ and subsequently many workers have extended both the basic theory (Gurtin et al., 1998; Huang and Sun, 2007; Huang and Wang, 2006; Steigmann and Ogden, 1999) as well examined its ramifications in various contexts e.g. nano-inclusions and effective properties of composites (Duan et al., 2005a, 2005b; He and Li, 2006; Lim et al., 2006; Mi and Kouris, 2014, 2007; Sharma and Ganti, 2004; Sharma et al., 2003; Sharma and Wheeler, 2007; Tian and Rajapakse, 2007a, 2007b), electronic structure of quantum dots (Sharma et al., 2003), mechanical response of nanostructures (Gupta and Kumar, 2018; He et al., 2004; Lim and He, 2004; Miller and Shenoy, 2000; Qu and Cherkaoui, 2007), grain boundary sliding (Weissmüller et al., 2011), response of rough surfaces (Duan et al., 2009; Mohammadi et al., 2013; Mohammadi and Sharma, 2012; Weissmüller and Duan, 2008), wrinkling (Fried and Todres, 2005), and sensors and resonators (Duan et al., 2009; Park and Klein, 2008). The literature on capillary effects is vast indeed and we do not make any attempt to be exhaustive in our review. We simply refer the reader to the following expositions that provide a guide to the pertinent literature albeit the tone of the reviews is understandably influenced by the interest of the authors: Müller and Saúl (2004). Cammarata (2009), Wang et al. (2011), and Javili et al. (2013). We specifically highlight a very informative and relevant review by Style et al. (2017) which specifically focuses on soft solids.

Loosely speaking, the deformation-dependent surface energy for solids may be expanded around a suitable reference state and considered to scale as⁶: $\gamma_0 + \tau_0 \cdot \epsilon_s + \frac{1}{2}E_s \cdot \epsilon_s^2$ + higher order terms. Here, ϵ_s is a suitable measure of the surface strain and γ_0 is a trivial constant. The so-called surface stress then scales as $\tau_0 + E_s \cdot \epsilon_s$ and is the key physical quantity of interest. The first multiplier of the strain (τ_0) is the residual surface stress that exists even in complete absence of an external strain. For an isotropic solid surface this quantity is a scalar and perhaps due to this reason, in analogy with liquids, it is sometimes (erroneously) referred to as the surface tension.⁷ The term E_s is a measure of the surface *elasticity* and parametrizes the change in surface stress due to deformation. For crystalline surfaces, τ_o is of the order of 1 N/m (Miller and Shenoy, 2000) and surface elastic stiffness is about an order of magnitude larger. For simple liquids and soft matter, τ_o is of the order of 1 mN/m (Style et al., 2015a). Conventional wisdom is that residual stresses do not alter the overall effective elastic behavior of composites. Therefore, incorporation of surface elasticity is essential to assess the impact of capillarity on overall elastic response of composites. As an example, we cite the recent work by Chatzigeorgiou et al. (2017) which specifically highlights neglecting the interface tension and focuses only on surface elasticity. Several works however have appeared that relate, what is apparently, the residual stress to the overall effective properties and surface elasticity is, at least prima facie, ignored-specifically, this certainly appears to be the case for Style et al. which successfully explains the experimental observations (Style et al., 2015a, 2015b). As we will outline in due course, the central issue underpinning these observations pertains to which framework (Eulerian versus Lagrangian) is used, what reference state is utilized for the even-

³ The length scale at which the surface-to-volume ratio becomes comparable to a characteristic length scale dictated by the strength of surface properties, indicates the size-scale when surface energy effects become important. Such a capillary length scales with σ^s/E where σ^s is a measure of the surface stress while *E* is indicative of the bulk elastic stiffness

⁴ An unfortunate consequence of the parallel developments in multiple areas is that at least in some cases, the same word may not necessarily describe the precisely the same physical context—and this is one aspect we will highlight in this work.

⁵ We remark that the Gurtin–Murdoch paper received relatively little attention until nearly a full two and a half decades after its publication when, due to the intense interest in nanotechnology, a preoccupation with nanoscale size-effects came to fore.

⁶ The discussion in this paragraph is intentionally somewhat heuristic, to establish some key points, and is restricted to the purely mechanical case.

⁷ For a simple liquid, the surface energy, surface stress and surface tension are sometimes interchangeably used in the literature, but this is not the case for solids.

tual linearization and even the postulate of the existence of a "stress-free" state.⁸ These subtleties are of lesser importance in hard solids but are quite relevant for highly deformable soft solids. We acknowledge here that these issues appear to be well-understood by some researchers e.g. Huang and Wang (2006, 2010, 2013), Wang et al. (2010) and Javili et al. (2018).

Germane to the central quest of the present work, several researchers have solved the Eshelby's inclusion problem within the Gurtin–Murdoch framework as well as addressed effective size-dependent properties of composites—the key literature on this topic was cited earlier. As an example, one of the present authors addressed this problem for spherical inclusions (Sharma and Ganti, 2004; Sharma et al., 2003) where the effective size-dependent bulk modulus was computed. In Sharma and Ganti (2004) a proposition analogous to Eshelby's was made that if surface energy is incorporated, only inclusions that have a constant *curvature* are capable of exhibiting a uniform elastic state under certain highly symmetric loading conditions. Duan et al. generalized the work on Eshelby's inclusion problem with surface energy beyond just the radially symmetric problem, and also discussed effective properties (Duan et al., 2005a, 2005b). More recently, in addition to the notable experiments mentioned in the topical paragraph of this article, Style and co-workers have also solved the problem of an incompressible liquid inclusion embedded in a soft solid (Style et al., 2015b). Using various levels of homogenization approximation (e.g. Mori and Tanaka, 1973), they successfully explained the experimental observations mentioned earlier. While we agree with the final result of the work by Style and co-workers—for the situation when a liquid inclusion is embedded in a soft incompressible solid—we believe that there are several subtleties related to, both the theories that purport to describe surface energy effects, as well as the solution to the liquid inclusion problem. In this work, we address the following subjects:

- (i) We advocate the viewpoint that while the Gurtin and Murdoch (1975) framework is quite general, its linearized version (as is typically interpreted and used in the literature) is more suitable for hard solids and that a somewhat altered theory is necessary for soft matter that are, in some sense, more "liquid like" yet still posses an apparent non-trivial surface elasticity.
- (ii) For capillarity in soft solids, we show that a "surface tension" like term gives rise to an *apparent* surface elasticity tensor that is distinct from the surface elasticity tensor of the Gurtin–Murdoch theory that is likely used in the context of hard solids. We believe that confusion regarding this aspect (although implicit) is prevalent in the literature and we elaborate regarding how considerable care must be taken before using or mapping the experimentally (or atomistically) determined surface elasticity theory with the appropriate continuum framework.
- (iii) We clarify the role of residual surface stresses on the effective elastic response of composites and the link between the residual surface stress and superficial elasticity.
- (iv) Mechanical stiffening effects due to capillary are of interest from both a fundamental scientific viewpoint as well as applications. However, perhaps an interesting application avenue is to exploit is the interplay between *electrostatics*, capillarity and liquid inclusions. This may be simply achieved by using ionic liquids instead of "ordinary" fluids.⁹ We extend the theoretical framework to incorporate electrical effects and consider the impact of using ionic liquid inclusions on the overall electrostriction and explore the ramifications on effective electrostriction and energy conversion. The motivation to develop soft materials with a strong electromechanical coupling can hardly be overstated. Applications range from soft robots to biomedical devices (Brochu and Pei, 2010; Carpi et al., 2011; Carpi and Smela, 2009; Lu and Kim, 2014; Shepherd et al., 2011; Shian et al., 2015). Given the extensive recent activity on this topic, for appropriate context, we simply point the reader toward the following articles (and references therein) that have addressed both the basic theory (Dorfmann and Ogden, 2005; Liu, 2013, 2014; McMeeking and Landis, 2005; Suo, 2010; Suo et al., 2008; Xiao and Bhattacharya, 2008) as well as the prediction of effective electromechanical coupling in soft matter (Hakimi Siboni and Ponte Castañeda, 2013; Lefevre and Lopez-Pamies, 2014; Liu and Sharma, 2018; Spinelli et al., 2015).

The outline of the paper is as follows: in Section 2 we establish the theoretical framework for surface energy in soft solids and contrast it with the typically used version of Gurtin–Murduch approach. In Section 3, we derive the effective mechanical properties of soft composites with liquid inclusions and capillary effects. We compare our results with those of Style et al. in Section 4, and extend the consideration to the electromechanical case by discussing the effective electrostrictive properties of soft composites and discuss pertinent insights related to exploiting the size-dependent interplay between capillarity and liquid inclusions for energy conversion.

Notation. For brevity, wherever possible, we employ direct notation. Vectors and tensors are denoted by bold symbols such as **u**, **I**, etc. When index notation is used, the convention of summation over repeated indices is followed. The tensor product between two vectors $\mathbf{a}, \mathbf{b} \in \mathbb{R}^3$ is defined as $(\mathbf{a} \otimes \mathbf{b})_{ij} = (\mathbf{a})_i(\mathbf{b})_j$ whereas the inner (or dot) product is defined as $\langle \mathbf{a}, \mathbf{b} \rangle \equiv \mathbf{a} \cdot \mathbf{b} := (\mathbf{a})_i(\mathbf{b})_i$, and the inner (or dot) product between matrices **A** and **B** of the same size is defined as $\mathbf{A} \cdot \mathbf{B} := \text{Tr}(\mathbf{A}^T \mathbf{B}) = (\mathbf{A})_{ij}(\mathbf{B})_{ij}$. For a domain D, $f_D(\cdot)$ denote the average of the integrand on D. From the viewpoint of matrices, the *i*th row vector of the gradient of a vector field, e.g., $\nabla \mathbf{u}$, is the gradient of the *i*th component of \mathbf{u} (with respect the Lagrangian coordinates \mathbf{x} , unless stated otherwise) whereas the "div" operates on the row vectors of a matrix field. Therefore,

⁸ In some scientific communities, these notions are not explicitly noted which is perhaps one reason that confusion may arise if equations are "transferred" from one community to the other without appropriate discussion or modification.

⁹ We remark that indeed, in the experiments conducted by Style et al. (2015a) the liquid inclusions were ionic in nature although exploration of electromechanical coupling was not the focus of their work.



Fig. 2. S_0 is a smooth surface oriented by a choice of unit normal field $\mathbf{n}(\mathbf{x})$.

div ∇ **u** = Δ **u** and div $[(\nabla$ **u**)^{*T*}] = ∇ (div**u**). For a scaling parameter $0 < \varepsilon \ll 1$ and a real number *n*, $O(\varepsilon^n)$ implies the asymptotic behavior $O(\varepsilon^n)/\varepsilon^n \to C \neq 0$ as $\varepsilon \to 0$ whereas $o(\varepsilon^n)/\varepsilon^n \to 0$ as $\varepsilon \to 0$.

2. Theoretical framework for surface energy in soft solids, comparison with the commonly used interpretation of the Gurtin–Murdoch model and associated subtleties

After summarizing the preliminary notions regarding surface kinematics and differential geometry in Section 2.1, we articulate the continuum theory for elastic material surfaces and associated linearization in Section 2.2. We discuss the associated subtleties germane to the present work in Section 2.3.

2.1. Surface kinematics

Consider a deformable material surface as sketched in Fig. 2 with a reference configuration $S_0 \subset \mathbb{R}^3$. For future convenience, we introduce a few notions. On S_0 and for a fixed material point, let **n** be the unit normal on S_0 , $\mathbf{I}_s = \mathbf{I} - \mathbf{n} \otimes \mathbf{n}$ the identity tensor on the tangent plane $\mathcal{T} := \{\mathbf{e} \in \mathbb{R}^3 : \mathbf{e} \cdot \mathbf{n} = 0\}$, $\mathbb{S} = \{\mathbf{M} \in \mathbb{R}^{3 \times 3} : \mathbf{Mn} = 0\}$ be an **n**-dependent subspace of $\mathbb{R}^{3 \times 3}$, $\mathbb{S}_{sym} = \{\mathbf{M} \in \mathbb{S} : \mathbf{M}^T = \mathbf{M}\}$, and $\mathbb{P} : \mathbb{R}^{3 \times 3} \to \mathbb{S}_{sym}$ be the orthogonal projection. We remark that these notions (i.e., \mathbf{n}, \mathbf{I}_s , $\mathcal{T}, \mathbb{P}, \mathbb{S}, \mathbb{S}_{sym}$) depend on the particular material point $\mathbf{x} \in S_0$ for a curved surface. For brevity, we subsequently omit the \mathbf{x} -dependence in notation.

Suppose that a scalar field φ and a vector field **u** are differentiable up to the boundary S_0 . The surface gradient ∇_s is defined for any scalar differentiable function φ by the following¹⁰:

$$\nabla_{\mathbf{s}}\varphi = \nabla\varphi - (\nabla\varphi \cdot \mathbf{n})\mathbf{n} = \mathbf{I}_{\mathbf{s}}\nabla\varphi.$$

We denote by

$$u_n = \mathbf{u} \cdot \mathbf{n}, \qquad \mathbf{u}_s = \mathbf{u} - u_n \mathbf{n}$$

the normal and tangential components of the vector field \mathbf{u} on S_0 . It is also convenient to introduce the surface gradient and the surface divergence of the vector field \mathbf{u} which are respectively defined as

$$\nabla_{\mathbf{s}} \mathbf{u} = \nabla \mathbf{u} - (\nabla \mathbf{u})(\mathbf{n} \otimes \mathbf{n}), \qquad \nabla_{\mathbf{s}} \cdot \mathbf{u} = \operatorname{Tr}(\nabla_{\mathbf{s}} \mathbf{u}).$$
(2.2)

(2.1)

The following identities differential geometry will be found useful, see e.g. the textbook of Kühnel (2015):

$$\begin{cases} \nabla_{s}\mathbf{u} = \nabla_{s}\mathbf{u}_{s} + \mathbf{n} \otimes \nabla_{s}u_{n} + u_{n}\mathbf{L}, \\ \nabla_{s}\mathbf{u}_{s} = \mathbf{I}_{s}(\nabla_{s}\mathbf{u}_{s}) - \mathbf{n} \otimes (\mathbf{L}\mathbf{u}_{s}), \\ \operatorname{div}_{s}\mathbf{u} = \operatorname{Tr}(\nabla_{s}\mathbf{u}) = \operatorname{div}_{s}\mathbf{u}_{s} + 2Hu_{n}, \end{cases}$$
(2.3)

where $\mathbf{L} = \nabla_s \mathbf{n} : \mathcal{T} \to \mathcal{T}$ is the Weingarten map (shape operator) on the tangential plane of the reference surface Ω_0 , and $H = \frac{1}{2} \text{Tr} \mathbf{L}$ is the mean curvature.

2.2. Surface energy, stress and elasticity

Consider a deformable body with reference configuration $\mathcal{B} \subset \mathbb{R}^3$ with \mathcal{S}_0 being its boundary. Let $\mathbf{y} : \mathcal{B} \to \mathbb{R}^3$ be the (continuously differentiable up to the boundary) deformation and as usual, denote by

$$\mathbf{F} = \nabla \mathbf{y}, \quad J = \det \mathbf{F}, \quad \mathbf{C} = \mathbf{F}^T \mathbf{F}$$
(2.4)

the deformation gradient, Jacobian, and Cauchy–Green tensor, respectively. The elastic energy of the system may be written as

$$F[\mathbf{y}] = E_s[\mathbf{y}] + \int_{\mathcal{B}} W(\nabla \mathbf{y})$$
(2.5)

¹⁰ By the convention employed in this work, the surface gradient at a fixed point $\nabla_s \mathbf{u} : \mathcal{T} \to \mathbb{R}^3$ for a vector field $\mathbf{u} : S_0 \to \mathbb{R}^3$ can be regarded as a matrix with the vanishing last column (instead of row).

where the first term arises from the elasticity of the material surface¹¹ and the second term from the bulk elasticity. Initial works on surface energy have a strong focus on "fluidic surfaces" where the elastic energy of the surface is typically defined for the current configuration and independent of the reference configuration. For example, the capillary model or constant surface tension model assumes that the elastic energy of the material surface is given by¹²

$$E_{s}[\mathcal{S}] = \int_{\mathcal{S}} \gamma \, ds, \tag{2.6}$$

where γ is conventionally referred to as the "surface tension".

From the viewpoint of nonlinear elasticity, Gurtin and Murdoch¹³ established a general elastic theory for material surfaces. The theory can be equivalently formulated by postulating the elastic energy of the surface can be written as a functional of surface deformation gradient¹⁴:

$$E_{s}[\mathbf{y}] = \int_{\mathcal{S}_{0}} W_{s}(\nabla_{s}\mathbf{y}) ds_{0}, \qquad (2.7)$$

where $W_s : \mathbb{R}^{3 \times 3} \to \mathbb{R}$ is the surface elastic energy density (per unit area of the reference surface). The surface Piola-Kirchhoff stress is defined as

$$\boldsymbol{\sigma}_{s} := \frac{\partial W_{s}(\mathbf{F}_{s})}{\partial \mathbf{F}_{s}} \qquad (\mathbf{F}_{s} = \nabla_{s} \mathbf{y}).$$
(2.8)

As for the classical 3D elasticity, the frame-indifference implies that the surface energy density $W_s : \mathbb{R}^{3\times 3} \to \mathbb{R}$ necessarily satisfies that for any rigid rotation $\mathbf{R} \in SO(3)$,

$$W_s(\mathbf{RF}) = W_s(\mathbf{F}) \qquad \forall \mathbf{F} \in \mathbb{R}^{3 \times 3}.$$

Therefore, the surface energy density can be rewritten as a function of the (surface) Cauchy-Green tensor:

$$W_{s}(\mathbf{F}) = W_{s}(\mathbf{U}_{s}), \qquad \mathbf{U}_{s} = (\mathbf{C}_{s})^{1/2}, \qquad \mathbf{C}_{s} = (\nabla_{s}\mathbf{y})^{T} \nabla_{s}\mathbf{y} \in \mathbb{S}_{sym},$$

where $\mathbf{C}_s : \mathcal{T} \to \mathcal{T}$ measures the deformation of the material surface S_0 . In particular, an infinitesimal line segment of length dl_0 along the direction $\mathbf{e}_0 \in \mathcal{T}$ on the reference configuration S_0 would have length $dl = (\mathbf{e}_0 \cdot \mathbf{C}_s \mathbf{e}_0)^{1/2} dl_0$ after the deformation. Moreover, the tensor \mathbf{C}_s can be represented as a 2 × 2 symmetric matrix if an orthonormal basis of the tangent space \mathcal{T} is fixed.

For practical applications, it is often necessary to linearize the general Gurtin–Murdoch model (2.7). Here we briefly articulate the linearized model conventionally used in the literature. We emphasize again that the original Gurtin–Murdoch framework is quite general however its interpretation and use in the literature is what perhaps can lead to confusion and we will make this statement more precise in this and Section 2.3. Following the familiar procedure in continuum mechanics, we introduce displacement $\mathbf{u}(\mathbf{x}) = \mathbf{y}(\mathbf{x}) - \mathbf{x}$ and assume small strain $|\nabla_s \mathbf{u}| \sim \varepsilon \ll 1$. Then we have

$$\mathbf{C}_{s} = ((\nabla_{s}\mathbf{x})^{T} + (\nabla_{s}\mathbf{u})^{T})(\nabla_{s}\mathbf{x} + \nabla_{s}\mathbf{u}) = \mathbf{I}_{s} + 2\mathbf{E}_{s} + (\nabla_{s}\mathbf{u})^{T}\nabla_{s}\mathbf{u},$$
(2.9)

where $\mathbf{E}_{s} \in \mathbb{S}_{sym}$, given by

$$\mathbf{E}_{s} = \frac{1}{2} \mathbb{P}[(\nabla_{s} \mathbf{u}) + (\nabla_{s} \mathbf{u})^{T}], \qquad (2.10)$$

is the *surface* symmetrized linear strain (cf., GM 1975, Eq. (3.6)). Then the surface energy density function can be expanded and truncated as:

$$\hat{W}_{s} = \hat{W}_{s}(\mathbf{U}_{s}) = \hat{W}([\mathbf{I}_{s} + 2\mathbf{E}_{s} + (\nabla_{s}\mathbf{u})^{T}\nabla_{s}\mathbf{u}]^{1/2}) = \hat{W}_{s}\left(\mathbf{I}_{s} + \mathbf{E}_{s} + \frac{1}{2}(\nabla_{s}\mathbf{u})^{T}\nabla_{s}\mathbf{u} - \frac{1}{2}\mathbf{E}_{s}^{2} + o(\varepsilon^{2})\right)$$
$$= \hat{W}_{s}(\mathbf{I}_{s}) + \boldsymbol{\sigma}_{s}^{0} \cdot \left[\mathbf{E}_{s} + \frac{1}{2}(\nabla_{s}\mathbf{u})^{T}\nabla_{s}\mathbf{u} - \frac{1}{2}\mathbf{E}_{s}^{2}\right] + \frac{1}{2}\mathbf{E}_{s} \cdot \hat{\mathbb{C}}_{s}\mathbf{E}_{s} + o(\varepsilon^{2}), \qquad (2.11)$$

where

$$\sigma_{s}^{0} = \frac{\partial \hat{W}_{s}(\mathbf{U}_{s})}{\partial \mathbf{U}_{s}} \bigg|_{\mathbf{U}_{s}=\mathbf{I}_{s}} \quad \text{and} \quad \hat{\mathbb{C}}_{s} = \frac{\partial^{2} \hat{W}_{s}(\mathbf{U}_{s})}{\partial \mathbf{U}_{s} \partial \mathbf{U}_{s}} \bigg|_{\mathbf{U}_{s}=\mathbf{I}_{s}}$$
(2.12)

can be interpreted as *surface* residual stress and *surface* elasticity tensor, respectively. In a standard linearization approach, it is tacitly assumed that the reference configuration is critical and stress-free as shown in Fig. 3(a), i.e., $\sigma_s^0 = 0$, or close to

¹¹ In what follows, we assume that the surface adheres to the bulk without "slipping". More general kinematics which allow interfacial jumps, incoherency and related situations were addressed by Gurtin et al. (1998) and Chatzigeorgiou et al. (2017).

¹² Another widely-used elastic model for fluidic membranes is the Helfrich model with energy given by $E_s[S] = \int_S 2\kappa_b H^2 + \kappa_g G ds$, where κ_b , κ_g are the bending moduli, and H (resp. G) is the local mean (resp. Gaussian) curvature of the surface S.

¹³ The original Gurtin–Murdoch theory used stress as the primitive concept while our preference here is to use surface energy as the starting point.

¹⁴ We do remark however that the Gurtin and Murdoch did not consider *curvature*-dependent (or $\nabla_s \nabla_s \mathbf{y}$ -dependent) surface energy which can be important under certain situations. This point was first raised by Steigmann and Ogden (1999) and later pursued in various applications by Fried and Todres (2005) and Chhapadia et al. (2011).



Fig. 3. (a) Linearization of the Gurtin–Murdoch model as it is typically used in the literature. (b) Linearized capillary model ($\epsilon \sim |\nabla_s \mathbf{u}|$).

critical in the sense that

$$\boldsymbol{\sigma}_{s}^{0} = \frac{\partial \widehat{W}_{s}(\mathbf{U}_{s})}{\partial \mathbf{U}_{s}}\Big|_{\mathbf{U}_{s}=\mathbf{I}_{s}} \sim O(\varepsilon).$$
(2.13)

Then, upon neglecting all higher order terms $o(\varepsilon^2)$, by (2.11) the surface energy density function can be approximated as

$$\hat{W}_{s} = \hat{W}_{s}(\mathbf{I}_{s}) + \boldsymbol{\sigma}_{s}^{0} \cdot \mathbf{E}_{s} + \frac{1}{2}\mathbf{E}_{s} \cdot \hat{\mathbb{C}}_{s}\mathbf{E}_{s} + o(\varepsilon^{2}).$$
(2.14)

From the definition (2.12), it is clear that the surface residual stress σ_s^0 necessarily belongs to \mathbb{S}_{sym} , and the surface stiffness tensor $\hat{\mathbb{C}}_s : \mathbb{S}_{sym} \to \mathbb{S}_{sym}$ possesses the properties: (i) the major symmetry: $\mathbf{M}_1 \cdot \hat{\mathbb{C}}_s \mathbf{M}_2 = \mathbf{M}_2 \cdot \hat{\mathbb{C}}_s \mathbf{M}_1$ for any $\mathbf{M}_1, \mathbf{M}_2 \in \mathbb{S}_{sym}$, (ii) the minor symmetry:

$$\hat{\mathbb{C}}_{s}\mathbf{M} = 0 \text{ if } \mathbf{M}^{T} = -\mathbf{M} \in \mathbb{S}, \tag{2.15}$$

and (iii) the positive-definiteness: $\mathbf{M} \cdot \hat{\mathbb{C}}_s \mathbf{M} > 0$ for any $0 \neq \mathbf{M} \in \mathbb{S}_{sym}$ if the free, standing-alone material surface S_0 is stable.¹⁵ Moreover, if the surface is isotropic in the sense that $\hat{W}_s(\mathbf{Q}^T \mathbf{U}_s \mathbf{Q}) = \hat{W}_s(\mathbf{U}_s)$ for any $\mathbf{U}_s \in \mathbb{S}_{sym}$ and rigid rotation $\mathbf{Q} \in SO(2)$, then it can be shown that the surface stress has to be given by

$$\boldsymbol{\sigma}_{s} = \boldsymbol{\sigma}_{s}^{0} + \hat{\mathbb{C}}_{s} \mathbf{E}_{s} = \boldsymbol{\sigma}_{s}^{0} \mathbf{I}_{s} + 2\mu_{s} \mathbf{E}_{s} + \lambda_{s} \mathbf{I}_{s} \mathrm{Tr}(\mathbf{E}_{s}), \quad \boldsymbol{\sigma}_{s}^{0} = \boldsymbol{\sigma}_{s}^{0} \mathbf{I}_{s}, \quad \hat{\mathbb{C}}_{s} \mathbf{E}_{s} = 2\mu_{s} \mathbf{E}_{s} + \lambda_{s} \mathbf{I}_{s} \mathrm{Tr}(\mathbf{E}_{s}), \quad (2.16)$$

where σ_s^0 is the isotropic residual surface stress, and (μ_s, λ_s) is the analogous Lamé constants.

Though primarily applied to solids by practitioners, the *nonlinear* Gurtin–Murdoch's formulation (2.7) is quite general, including the capillary model (2.6) for fluidic surfaces as a special case. To see this, by a change of variables we can rewrite the capillary energy (2.6) as a functional of deformation:

$$E_{s}[\mathbf{y}] = \int_{\mathcal{S}_{0}} \gamma \{ \det[(\nabla_{s}\mathbf{y})^{T}\nabla_{s}\mathbf{y}] \}^{1/2} ds_{0} = \int_{\mathcal{S}_{0}} \hat{W}_{s}(\mathbf{U}_{s}) ds_{0}, \qquad \hat{W}_{s} = \gamma \det \mathbf{U}_{s}.$$
(2.17)

It is also clear that the capillary model is isotropic: $\hat{W}_s(\mathbf{Q}^T \mathbf{U}_s \mathbf{Q}) = \hat{W}_s(\mathbf{U}_s)$ for any $\mathbf{Q} \in SO(2)$. According to the above argument from (2.9) to (2.16), for a linearized capillary model one might anticipate a truncated energy form specified by (2.14) and (2.16). It turns out, somewhat surprisingly, that the linearized capillary model admits quite a different form of surface stiffness tensor. To see this, by (2.12) and direct calculation we find that (2.14) can be written as

$$\hat{W}_{s} = \gamma + \gamma \operatorname{Tr} \mathbf{E}_{s} + \frac{\gamma}{2} [(\operatorname{Tr} \mathbf{E}_{s})^{2} - |\mathbf{E}_{s}|^{2}] + o(\varepsilon^{2}),$$
(2.18)

and hence by (2.8), the (linearized) Piola-Kirchhoff stress is given by

$$\sigma_s = \gamma \mathbf{I}_s - \gamma \mathbf{E}_s + \gamma \mathbf{I}_s \mathrm{Tr} \mathbf{E}_s. \tag{2.19}$$

However, if the calculation in (2.11)–(2.14) were carefully repeated for the particular surface energy density $\hat{W}_s = \gamma \det \mathbf{U}_s$, one would find that (2.14) is no longer valid and by (2.11), should be replaced by

$$\hat{W}_{s} = \hat{W}_{s}(\mathbf{I}_{s}) + \gamma \operatorname{Tr} \mathbf{E}_{s} + \frac{\gamma}{2} \operatorname{Tr}[(\nabla_{s} \mathbf{u})^{T} \nabla_{s} \mathbf{u} - \mathbf{E}_{s}^{2}] + \frac{\gamma}{2} [(\operatorname{Tr} \mathbf{E}_{s})^{2} - |\mathbf{E}_{s}|^{2}] + o(\varepsilon^{2}),$$
(2.20)

where the additional quadratic term $\frac{\gamma}{2}$ Tr[($\nabla_s \mathbf{u}$)^T $\nabla_s \mathbf{u} - \mathbf{E}_s^2$] arises from the fact that the surface residual stress σ_s^0 is of order O(1) instead of $O(\varepsilon)$ (cf., (2.13)). In other words, whenever the reference configuration S_0 is not close to the critical "stress-free" configuration as shown in Fig. 3(b) (i.e., the surface strain \mathbf{U}_s such that $\partial \hat{W}_s(\mathbf{U}_s)/\partial \mathbf{U}_s = 0$), there is additional quadratic

¹⁵ In solids, the material surface S_0 usually represents the interface between two different phases and does not "stand alone". Therefore, the positivedefiniteness of $\hat{\mathbb{C}}_s$ can be violated, as shown in atomistic simulations (e.g. Hu and Liu, 2015; Shenoy, 2005).

contributions from the "residual stress" in the expansion of energy function that is important for the correct linearization of a nonlinear surface elasticity theory. The same phenomenon can occur in 3D nonlinear elasticity, though it is rare that the reference configuration in 3D elasticity is chosen not to be close to a stress-free configuration (i.e., (2.13) is violated). For the capillary model $\hat{W}_s = \gamma \det \mathbf{U}_s$, a stress-free configuration is a surface with zero area which is apparently not close to the reference configuration S_0 . This gives rise to the extra terms in (2.20) as compared with (2.18).

2.3. A simplified summary and discussion of the key points underpinning surface elasticity as relevant for soft solids

With the benefit of the mathematical derivations of the linearized surface elasticity theory articulated in the previous section, we attempt now to discuss some of the pertinent nuances for soft solids. The fluid mechanics community typically work in the current configuration and invoke the surface energy in the form (2.6). Gurtin and Murdoch, in targeting solids, advocated to separate the superficial elasticity of solid from the surface tension (2.6) and appeared to exclusively refer to the "surface elasticity tensor" Gurtin and Murdoch (1975) being contributed by a solid surface. They also showed that how two different but closely related surface elasticity tensors may emerge from the linearization procedure, depending on whether one chooses to expand and truncate the Piola-Kirchhoff stress tensor or the (pulled-back) Cauchy stress. From the viewpoint of energy, this amounts to starting with the surface energy functional with separate contributions from the surface tension and "solid" surface elasticity:

$$E_{s}[\mathbf{y}] = \int_{\mathcal{S}_{0}} [\gamma \det \mathbf{U}_{s} + \hat{W}_{s}(\mathbf{U}_{s})] ds_{0}, \qquad (2.21)$$

where \hat{W}_s is assumed to satisfy (2.13). Upon repeating the calculation from (2.11) to (2.20), we find that, for an isotropic surface the (linearized) total surface stress is given by

$$\boldsymbol{\sigma}_{s} = (\sigma_{s}^{0} + \gamma)\mathbf{I}_{s} + 2(\mu_{s} - \gamma)\mathbf{E}_{s} + (\lambda_{s} + \gamma)\mathrm{Tr}\mathbf{E}_{s} + \gamma\nabla_{s}\mathbf{u} =: \sigma_{0}\mathbf{I}_{s} + 2\mu_{0}\mathbf{E}_{s} + \lambda_{0}\mathrm{Tr}\mathbf{E}_{s} + \gamma\nabla_{s}\mathbf{u},$$
(2.22)

where σ_s^0 , μ_s , λ_s are defined by (2.12) and (2.16), $\sigma_0 = \sigma_s^0 + \gamma$, $\mu_0 = \mu_s - \gamma$, $\lambda_0 = \lambda_s + \gamma$, and the extra terms (compared with (2.16)) is contributed by the surface tension (c.f., (2.20)). For *hard* solids, it is reasonable to assume that the deformed state is close to the undeformed one and that there exists a stress-free state in its neighborhood. This justifies the omission of surface tension ($\gamma = 0$) and the assumption (2.13), and the typically used version of the Gurtin–Murdoch model where linearization is performed around such a suitable reference state with surface stress given by (2.16). In this case, the last term in (2.22) should be neglected as well for consistency and the (linearized) surface stress is *symmetric*. In a soft solid, which is closer to a fluid, neglecting the "solid" contribution with surface energy given by the first term in (2.21) alone i.e. (2.6) may be more reasonable. Where then does the apparent surface elasticity emerge from? The key observation is that even if the second term in (2.21) is neglected i.e. (λ_s , μ_s) = 0, (λ_0 , μ_0) in (2.22) do not vanish which gives rise to the apparent surface elasticity. In other words, the (linearized) Piola-Kirchhoff stress does depend on the deformation gradient or strain for a completely *deformation independent* surface tension and constant Cauchy stress in the current configuration.

In summary, we point out that the model in (2.6) is the prevailing minimum model to account for interfacial effects, particularly in the works concerning liquids (e.g. the Young-Laplace equation). The capillary model (2.6) in Eulerian form might appear to be simple, but is in fact nonlinear for the displacement **u** and the proper linearization yields the following truncated elastic energy functional for small strain $|\nabla_s \mathbf{u}| \sim \epsilon \ll 1$:

$$E_{s}[\mathbf{u}] = \int_{\partial\Omega_{0}} W_{s}(\nabla_{s}\mathbf{u}) ds_{0} = \int_{\partial\Omega_{0}} \left[\gamma + \gamma \mathbf{I}_{s} \cdot \nabla_{s}\mathbf{u} + \frac{1}{2}\nabla_{s}\mathbf{u} \cdot \mathbb{C}_{s}\nabla_{s}\mathbf{u} \right] ds_{0} + o(\epsilon^{2}), \qquad (2.23)$$

where, by (2.19) and (2.20), we have lumped all terms quadratic in $\nabla_s \mathbf{u}$ together by introducing the new "surface elasticity tensor" \mathbb{C}_s^{16} such that the surface stress $\boldsymbol{\sigma}_s$ is given by

$$\sigma_{s} = \gamma \mathbf{I}_{s} + \mathbb{C}_{s} \nabla_{s} \mathbf{u} = \gamma \mathbf{I}_{s} + \gamma [\nabla_{s} \mathbf{u} + (\nabla_{s} \cdot \mathbf{u})\mathbf{I}_{s} - \mathbf{I}_{s} (\nabla_{s} \mathbf{u}) - (\mathbf{I}_{s} \nabla_{s} \mathbf{u})^{\mathrm{T}}] = \gamma \mathbf{I}_{s} + \gamma (\mathbf{I}_{s} \mathrm{Tr} \mathbf{E}_{s} - 2\mathbf{E}_{s}) + \gamma \nabla_{s} \mathbf{u}.$$
(2.24)

We remark that as a linear mapping from S to S, the new surface elasticity tensor (2.24) associated with the capillary model does not enjoy the minor symmetry (2.15) and is not positive definite, and the surface stress σ_s is in general non-symmetric.

3. Effective elastic properties of soft composites with capillary effects

As illustrated in Fig. 4(b), we consider a composite model with a single inclusion Ω_0 embedded in a representative volume element $Y \subset \mathbb{R}^3$ of the composite. We choose the reference configuration to be the state with vanishing *bulk* stress in the matrix $Y \setminus \Omega_0$, and the solid or liquid inclusion $\Omega_0 \subset Y$ is assumed to be a sphere of radius R_0 in this reference configuration. A novel ingredient of present work lies in that the interface between the inclusion and matrix is an elastic entity and contributes an elastic energy that depends on the surface configuration. We describe the state of the composite by the displacement $\mathbf{u} = \mathbf{y} - \mathbf{x}$, where $\mathbf{y} : Y \to \mathbb{R}^3$ is the deformation (continuously differentiable up to the boundary $\partial \Omega_0$) and for simplicity, restrict ourselves to the regime of small strain¹⁷ (i.e., $|\nabla \mathbf{u}| \sim \varepsilon \ll 1$).

 $^{^{16}}$ The reader is cautioned that \mathbb{C}_s is different from the surface elasticity tensors Gurtin and Murdoch referred to in their works.

¹⁷ The existence of such a reference configuration is a postulation concerning the "mismatch" between the interface, interior bulk and exterior bulk.



Fig. 4. (a) Hashin's coated-sphere assemblage. (b) A simplified representative volume element of the composite consisting of three components: an exterior domain, a spherical inclusion, and an interface.

3.1. Surface elastic model for a spherical incompressible inclusion

Although the linearized Gurtin–Murdoch model of form (2.16) is widely applied to model interfaces in solids and (redblood) cellular membranes, in this work we, however, employ the capillary model or surface tension model (2.6). We remark that effective elastic properties of composites with a linearized Gurtin–Murdoch surface elasticity of form (2.16) have been addressed in the works of Duan et al. (2005b), Sharma et al. (2003) and others.

For simplicity, we employ the truncated surface energy density in (2.23) of the capillary model. For closed surfaces, there is a physically important scenario when the reference surface $\partial \Omega_0$ is spherical of radius R_0 and the interior volume remains to be constant. Such a model and reference configuration is natural for incompressible liquid or soft inclusions. In this case, the incompressibility constraint implies that

$$1 = \frac{1}{\operatorname{vol}(\Omega_0)} \int_{\Omega_0} \det(\mathbf{I} + \nabla \mathbf{u}) d\nu_0 = 1 + \frac{1}{\operatorname{vol}(\Omega_0)} \int_{\Omega_0} \left(\nabla \cdot \mathbf{u} + \frac{1}{2} \nabla \mathbf{u} \cdot \mathbb{T} \nabla \mathbf{u} \right) ds_0 + o(\varepsilon^2), \tag{3.1}$$

where the fourth-order tensor $\mathbb{T}: \mathbb{R}^{3\times 3} \to \mathbb{R}^{3\times 3}$ is given by

$$\mathbb{T}\mathbf{F} = (\mathrm{T}\mathbf{r}\mathbf{F})\mathbf{I} - \mathbf{F}^{T} \quad \forall \mathbf{F} \in \mathbb{R}^{3\times3} \qquad \text{i.e.} \qquad (\mathbb{T})_{ijkl} = -\delta_{il}\delta_{jk} + \delta_{ij}\delta_{kl}.$$
(3.2)

By the divergence theorem, we find that

$$\int_{\partial\Omega_0} \mathbf{I}_s \cdot \nabla_s \mathbf{u} ds_0 = \int_{\partial\Omega_0} \frac{2}{R_0} u_n ds_0 = o(\varepsilon^2) - \frac{1}{2} \int_{\partial\Omega_0} \frac{2}{R_0} (\mathbf{u} \otimes \mathbf{n}) \cdot \mathbb{T} \nabla_s \mathbf{u} ds_0,$$
(3.3)

where the first equality follows from (2.3), and the second equality follows from (3.1). In other words, the second integral $\int_{\partial \Omega_0} \mathbf{I}_s \cdot \nabla_s \mathbf{u} ds_0$ in (2.23) in fact scales as $O(\varepsilon^2)$ —the same as the third term in (2.23). Upon eliminating the term $\int_{\Omega_0} \mathbf{I}_s \cdot \nabla_s \mathbf{u} ds_0$ by (3.3), we obtain the leading strain energy for constant-volume quasi-spherical surfaces associated with the capillary model (Liu et al., 2017):

$$E_{s}[\mathbf{u}] = \frac{1}{2} \int_{\partial \Omega_{0}} \left[\nabla_{s} \mathbf{u} \cdot \mathbb{C}_{s} \nabla_{s} \mathbf{u} - \frac{2\gamma}{R_{0}} (\mathbf{u} \otimes \mathbf{n}) \cdot \mathbb{T} \nabla_{s} \mathbf{u} \right] ds_{0},$$
(3.4)

where an additive constant has been neglected.

Remark 1. In the subsequent sections, we choose the expression (3.4) as the surface elastic energy for spherical interfaces with *incompressible* interior.¹⁸ As one will see shortly, the surface elasticity with incompressible interior in general "stiffens" the composite since (3.4) is positive for nonzero $|\nabla_s \mathbf{u}|$. However, the capillary effect can "soften" the composite if the inclusion is non-spherical or compressible for some applied loading direction since the tensor \mathbb{C}_s defined in (2.24) is not positive-definite.

¹⁸ Nevertheless, we subsequently present solutions to bulk elasticity for *compressible* interior with a bulk modulus κ^{in} for brevity and potential generalization. The reader is cautioned that the final results only apply to incompressible interior with $\kappa^{\text{in}} \to +\infty$.

3.2. Homogenization formulation

For simplicity, we assume that the composite has periodic microstructure with $Y = (0, 1)^3$ being a unit cell or representative volume element (RVE). The microstructure is illustrated in Fig. 4(b), consisting of exterior bulk $Y \setminus \Omega_0$, spherical interface $\partial \Omega_0$, and interior bulk Ω_0 . For the exterior and interior bulks, their strain energies are modeled by the linearized elasticity with a stiffness tensor \mathbb{C}^{ex} and \mathbb{C}^{in} , respectively. Then the total elastic energy per RVE can be written as

$$F_{1}[\mathbf{u}] = E_{s}[\mathbf{u}] + E_{b}^{\text{in}}[\mathbf{u}] + E_{b}^{\text{ex}}[\mathbf{u}] = E_{s}[\mathbf{u}] + \int_{Y} \left[\boldsymbol{\sigma}_{\text{res}} \cdot \nabla \mathbf{u} + \frac{1}{2} \nabla \mathbf{u} \cdot \mathbb{C}_{\#} \nabla \mathbf{u} \right],$$
(3.5)

where $E_b^{\text{in}}[\mathbf{u}]$ and $E_b^{\text{ex}}[\mathbf{u}]$ stand respectively for the elastic energy in the inclusion and the matrix, σ_{res} is the possible residual stress in the interior bulk, and the **x**-dependent bulk stiffness tensor $\mathbb{C}_{\#}$ is given by

$$\mathbb{C}_{\#} = \begin{cases} \mathbb{C}^{ex} & \text{in } Y \setminus \Omega_0, \\ \mathbb{C}^{in} & \text{in } \Omega_0. \end{cases}$$

We are interested in the effective properties of the composite incorporating the effects of surface elasticity. Let $\tilde{\mathbf{E}} \in \mathbb{R}_{sym}^{3\times 3}$ the prescribed average strain of the composite and $\mathcal{U}_{\tilde{\mathbf{F}}}$ be the collection of admissible displacements:

$$\mathcal{U}_{\bar{\mathbf{E}}} = \left\{ \mathbf{u} : \int_{Y} \nabla \mathbf{u} = \bar{\mathbf{E}} \text{ and } \nabla \mathbf{u} \text{ is } Y \text{-periodic} \right\}.$$
(3.6)

From the principle of minimum free energy, the equilibrium state of the composite is governed by the minimization problem:

$$\Psi_1^{\text{eff}}(\tilde{\mathbf{E}}) := \min\{F_1[\mathbf{u}]: \ \mathbf{u} \in \mathcal{U}_{\tilde{\mathbf{E}}}\},\tag{3.7}$$

where Ψ_1^{eff} : $\mathbb{R}_{\text{sym}}^{3\times3} \to \mathbb{R}$ is referred to as the effective energy density. By the standard first-variation calculation, the Euler–Lagrange equations associated with the variational principle (3.7) in the bulk are given by

$$\begin{aligned} &\operatorname{div}(\mathbb{C}^{\mathrm{ex}}\nabla\mathbf{u}) = 0 & \operatorname{in} \ Y \setminus \Omega_0, \\ &\operatorname{div}(\mathbb{C}^{\mathrm{in}}\nabla\mathbf{u}) = 0 & \operatorname{in} \ \Omega_0. \end{aligned}$$
 (3.8)

In analogy with the classic homogenization theory (Milton, 2002), we regard the overall composite as a "bulk" elastic material with an effective *bulk* stiffness tensor defined as

$$\mathbb{C}^{\text{eff}} = \frac{\partial^2 \Psi_1^{\text{eff}}}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{E}}} \Big|_{\bar{\mathbf{E}}=0}.$$
(3.9)

Since the energy function (3.5) is quadratic, we observe that \mathbb{C}^{eff} is independent of σ_{res} which is subsequently set to be zero.

Evaluating the effective properties of the composite amounts to solving the minimization problem (3.7). However, exactly solvable microstructures are rare; an exception includes the classic example of Hashin's assemblage of coated spheres (cf., Fig. 4(a), Hashin (1962)). Before proceeding to the detailed solution, it is convenient to rewrite the energy (3.5) as a surface integral while enforcing the bulk equilibrium Eq. (3.8)_{1,2}. Let $\mathbf{u} \in U_{\tilde{E}}$ be a trial solution satisfying (3.8)_{1,2}, \mathbf{u}' be the "astray" displacement:

$$\mathbf{u}' = \mathbf{u} - \bar{\mathbf{u}}, \qquad \bar{\mathbf{u}} = \bar{\mathbf{E}}\mathbf{x}. \tag{3.10}$$

We remark that \mathbf{u}' itself is Y-periodic. By the divergence theorem and $(3.8)_1$, we find the bulk elastic energy in the matrix as

$$E_{b}^{\text{ex}}[\mathbf{u}] = \int_{Y \setminus \Omega_{0}} \frac{1}{2} \nabla \mathbf{u} \cdot \mathbb{C}^{\text{ex}} \nabla \mathbf{u} = \int_{Y \setminus \Omega_{0}} \left[\frac{1}{2} \mathbf{\tilde{E}} \cdot \mathbb{C}^{\text{ex}} \mathbf{\tilde{E}} + \mathbf{\tilde{E}} \cdot \mathbb{C}^{\text{ex}} \nabla \mathbf{u}' + \frac{1}{2} \nabla \mathbf{u}' \cdot \mathbb{C}^{\text{ex}} \nabla \mathbf{u}' \right]$$
$$= \frac{1}{2} (1 - \nu_{f}) \mathbf{\tilde{E}} \cdot \mathbb{C}^{\text{ex}} \mathbf{\tilde{E}} - (\mathbb{C}^{\text{ex}} \mathbf{\tilde{E}}) \cdot \int_{\partial \Omega_{0}^{+}} \mathbf{u}' \otimes \mathbf{n} - \frac{1}{2} \int_{\partial \Omega_{0}^{+}} \mathbf{u}' \cdot (\mathbb{C}^{\text{ex}} \nabla \mathbf{u}') \mathbf{n},$$
(3.11)

where $v_f = vol(\Omega_0)/vol(Y)$ is the volume fraction of the inclusion phase. Similarly, by the divergence theorem and $(3.8)_2$ we rewrite the bulk elastic energy in the inclusion as

$$E_{b}^{\mathrm{in}}[\mathbf{u}] = \int_{\Omega_{0}} \frac{1}{2} \nabla \mathbf{u} \cdot \mathbb{C}^{\mathrm{in}} \nabla \mathbf{u} = \int_{\Omega_{0}} \left[\frac{1}{2} \mathbf{\tilde{E}} \cdot \mathbb{C}^{\mathrm{in}} \mathbf{\tilde{E}} + \mathbf{\tilde{E}} \cdot \mathbb{C}^{\mathrm{in}} \nabla \mathbf{u}' + \frac{1}{2} \nabla \mathbf{u}' \cdot \mathbb{C}^{\mathrm{in}} \nabla \mathbf{u}' \right]$$
$$= \frac{1}{2} \nu_{\mathrm{f}} \mathbf{\tilde{E}} \cdot \mathbb{C}^{\mathrm{in}} \mathbf{\tilde{E}} + (\mathbb{C}^{\mathrm{in}} \mathbf{\tilde{E}}) \cdot \int_{\partial \Omega_{0}^{-}} \mathbf{u}' \otimes \mathbf{n} + \frac{1}{2} \int_{\partial \Omega_{0}^{-}} \mathbf{u}' \cdot (\mathbb{C}^{\mathrm{in}} \nabla \mathbf{u}') \mathbf{n}.$$
(3.12)

From (3.11), (3.12), and (3.4), we see that the total energy $F_1[\mathbf{u}]$ defined by (3.5) can be rewritten as a surface integral on $\partial \Omega_0$ (Recall that \mathbf{u}' is continuous across $\partial \Omega_0$):

$$F_{1}[\mathbf{u}] = E_{s}[\mathbf{u}] + \frac{1}{2}(1 - \nu_{f})\bar{\mathbf{E}} \cdot \mathbb{C}^{\text{ex}}\bar{\mathbf{E}} + \frac{1}{2}\nu_{f}\bar{\mathbf{E}} \cdot \mathbb{C}^{\text{in}}\bar{\mathbf{E}} + (\mathbb{C}^{\text{in}}\bar{\mathbf{E}} - \mathbb{C}^{\text{ex}}\bar{\mathbf{E}}) \cdot \int_{\partial\Omega_{0}} \mathbf{u}' \otimes \mathbf{n} - \frac{1}{2}\int_{\partial\Omega_{0}^{+}} \mathbf{u}' \cdot (\mathbb{C}^{\text{ex}}\nabla\mathbf{u}')\mathbf{n} + \frac{1}{2}\int_{\partial\Omega_{0}^{-}} \mathbf{u}' \cdot (\mathbb{C}^{\text{in}}\nabla\mathbf{u}')\mathbf{n}.$$
(3.13)

This observation motivates our approach: instead of directly solving the Euler–Lagrange equations associated with (3.7), we come back to the minimization problem (3.7) with the energy functional (3.13) which is now given as an surface integral on $\partial \Omega_0$.

3.3. Exact solution for coated spheres and effective elastic properties

For explicit solutions, we consider a coated sphere in the assemblage as illustrated in Fig. 4(a) and assume that (i) the exterior and interior solids are isotropic with μ^{in} , κ^{in} (resp. μ^{ex} , κ^{ex}) being the shear modulus and bulk modulus of the inclusion (resp. matrix), and (ii) the average strain $\mathbf{\tilde{E}}$ is prescribed as

$$\mathbf{E} = a[\mathbf{e}_{x} \otimes \mathbf{e}_{x} + \mathbf{e}_{y} \otimes \mathbf{e}_{y}] + b\mathbf{e}_{z} \otimes \mathbf{e}_{z}, \tag{3.14}$$

where $(\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z)$ are the basis vectors for the Cartesian coordinate system. Immediately, the first line in (3.13) can be written as

$$\frac{1}{2}(1-\nu_{\rm f})\bar{\mathbf{E}}\cdot\mathbb{C}^{\rm ex}\bar{\mathbf{E}}+\frac{1}{2}\nu_{\rm f}\bar{\mathbf{E}}\cdot\mathbb{C}^{\rm in}\bar{\mathbf{E}}=\frac{1}{2}\bar{\mathbf{E}}\cdot[(1-\nu_{\rm f})\mathbb{C}^{\rm ex}+\nu_{\rm f}\mathbb{C}^{\rm in}]\bar{\mathbf{E}}.$$
(3.15)

Upon specifying the boundary displacement

$$\mathbf{u}' = \mathbf{u}'_b \qquad \text{on } \partial\Omega_0, \tag{3.16}$$

the differential Eq. (3.8)_{1,2} uniquely determine the "astray" displacement \mathbf{u}' in both the exterior region $Y \setminus \Omega_0$ and the interior region Ω_0 . Since the inclusion Ω_0 is a sphere of radius R_0 , we employ spherical coordinates (r, θ, ϕ) and vector spherical harmonics, and write the boundary value of \mathbf{u}' on $\partial \Omega_0$ as

$$\mathbf{u}_{b}^{\prime}(\theta) = \sum_{l=0}^{\infty} [X_{l}^{\prime} \mathbf{Y}_{l}(\theta) + \Theta_{l}^{\prime} \Psi_{l}(\theta)], \qquad (3.17)$$

where the vectorial axis-symmetric spherical harmonics are defined as (see Appendix A.1 for details)

$$\mathbf{Y}_{l}(\theta) = Y_{l}(\theta)\mathbf{e}_{r}, \qquad \mathbf{\Psi}_{l} = r\nabla Y_{l} = \mathbf{e}_{\theta}\frac{d}{d\theta}Y_{l}(\theta), \qquad (3.18)$$

and $(\mathbf{e}_r, \mathbf{e}_{\theta}, \mathbf{e}_{\phi})$ are the unit basis vectors associated with spherical coordinates (r, θ, ϕ) . By (A.6) we rewrite the average strain (3.14) as

$$\tilde{\mathbf{E}} = (a\sin^2\theta + b\cos^2\theta)\mathbf{e}_r \otimes \mathbf{e}_r + (a - b)\sin\theta\cos\theta(\mathbf{e}_r \otimes \mathbf{e}_\theta + \mathbf{e}_\theta \otimes \mathbf{e}_r) + (a\cos^2\theta + b\sin^2\theta)\mathbf{e}_\theta \otimes \mathbf{e}_\theta + a\mathbf{e}_\phi \otimes \mathbf{e}_\phi.$$
(3.19)

Consequently, from Appendix A.1, (A.9) the second line in (3.13) can be written as

$$(\mathbb{C}^{\text{in}}\bar{\mathbf{E}} - \mathbb{C}^{\text{ex}}\bar{\mathbf{E}}) \cdot \int_{\partial\Omega_0} \mathbf{u}' \otimes \mathbf{n} = \frac{2}{15} \sqrt{\pi} R_0^2 (15X_0'(2a+b)(\kappa^{\text{in}} - \kappa^{\text{ex}}) + 4\sqrt{5}(a-b)(\mu^{\text{ex}} - \mu^{\text{in}})(3\Theta_2' + X_2')),$$
(3.20)

where we notice that only modes l = 0, 2 survive in (3.20) since every component of (3.19) is at most a quadratic function of $\hat{\mathbf{x}} := \mathbf{x}/|\mathbf{x}|$. The interior and exterior bulk energy associated with the astray displacement \mathbf{u}' is determined by the boundary value problem: (3.8)_{1,2}, $\int_{\{r=R\}} \mathbf{u}' \otimes \mathbf{e}_r = 0$, and the boundary conditions (3.16) and (3.17). By the standard procedure of separation of variables, we find that (see details in Appendix A.2)

$$\frac{1}{2} \int_{\Omega_0} \nabla \mathbf{u}' \cdot \mathbb{C}^{\text{in}} \nabla \mathbf{u}' = \frac{1}{2} \int_{\partial \Omega_0^-} \mathbf{u}' \cdot (\mathbb{C}^{\text{in}} \nabla \mathbf{u}') \mathbf{n} = \sum_{l=0}^{\infty} \frac{1}{2} \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix} \cdot \mathbf{D}_l^{\text{in}} \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix},
\frac{1}{2} \int_{Y \setminus \Omega_0} \nabla \mathbf{u}' \cdot \mathbb{C}^{\text{ex}} \nabla \mathbf{u}' = -\frac{1}{2} \int_{\partial \Omega_0^+} \mathbf{u}' \cdot (\mathbb{C}^{\text{ex}} \nabla \mathbf{u}') \mathbf{n} = \sum_{l=0}^{\infty} \frac{1}{2} \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix} \cdot \mathbf{D}_l^{\text{ex}} \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix},$$
(3.21)

where the 2 × 2 matrices $\mathbf{D}_{l}^{\text{in}}$ and $\mathbf{D}_{l}^{\text{ex}}$ for the leading two modes l = 0 and l = 2 are given by

$$\begin{aligned} \mathbf{D}_{0}^{\text{in}} &= R_{0}\kappa^{\text{in}} \begin{bmatrix} 3 & 0\\ 0 & 0 \end{bmatrix}, \quad \mathbf{D}_{0}^{\text{ex}} &= \frac{R_{0}(4\mu^{\text{ex}} + 3\kappa^{\text{ex}}v_{f})}{1 - v_{f}} \begin{bmatrix} 1 & 0\\ 0 & 0 \end{bmatrix}, \\ \mathbf{D}_{2}^{\text{in}} &= \frac{R_{0}\mu^{\text{in}}}{6\kappa^{\text{in}} + 17\mu^{\text{in}}} \begin{bmatrix} 39\kappa^{\text{in}} + 16\mu^{\text{in}} & 18(-3\kappa^{\text{in}} + 2\mu^{\text{in}})\\ 18(-3\kappa^{\text{in}} + 2\mu^{\text{in}}) & 12(15\kappa^{\text{in}} + 11\mu^{\text{in}}) \end{bmatrix}, \\ \mathbf{D}_{2}^{\text{ex}} &= \frac{2\mu^{\text{ex}}R_{0}}{3(1 - v_{f})(\kappa^{\text{ex}} + 2\mu^{\text{ex}})} \begin{bmatrix} 9\kappa^{\text{ex}} + 16\mu^{\text{ex}} - 6v_{f}(\kappa^{\text{ex}} + 2\mu^{\text{ex}}) & 3(\kappa^{\text{ex}}(6v_{f} - 3) + 4\mu^{\text{ex}}(3v_{f} - 2))\\ 3(\kappa^{\text{ex}}(6v_{f} - 3) + 4\mu^{\text{ex}}(3v_{f} - 2)) & 9(5\kappa^{\text{ex}} + 8\mu^{\text{ex}} - 2v_{f}(\kappa^{\text{ex}} + 2\mu^{\text{ex}})) \end{bmatrix}. \end{aligned}$$
(3.22)

Further, the surface elastic energy can also be written as a quadratic form of $(X_l, \Theta_l) = (\bar{X}_l, \bar{\Theta}_l) + (X'_l, \Theta'_l)$. In particular, if the displacement $\bar{\mathbf{u}} = \bar{\mathbf{E}}\mathbf{x} = (ax_1, ax_2, bx_3)$ on $\partial \Omega_0$ can be written as

$$\bar{\mathbf{u}} = \bar{X}_0 \mathbf{Y}_0 + \bar{X}_2 \mathbf{Y}_2 + \bar{\Theta}_2 \mathbf{\Psi}_2,$$

where

$$\bar{X}_0 = \frac{2}{3}R_0\pi^{1/2}(2a+b), \quad \bar{X}_2 = -\frac{4}{15}R_0(5\pi)^{1/2}(a-b), \quad \bar{\Theta}_2 = -\frac{2}{15}R_0(5\pi)^{1/2}(a-b).$$
(3.23)

By (3.23) and (A.9), we can rewrite (3.20) as a cross-term between $(\bar{X}_l, \bar{\Theta}_l)$ and (X'_l, Θ'_l) :

$$\left(\mathbb{C}^{\mathrm{in}} \tilde{\mathbf{E}} - \mathbb{C}^{\mathrm{ex}} \tilde{\mathbf{E}} \right) \cdot \int_{\partial \Omega_0} \mathbf{u}' \otimes \mathbf{n} = \sum_{l=0,2} \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix} \cdot \mathbf{B}_l \begin{bmatrix} \bar{X}_l \\ \bar{\Theta}_l \end{bmatrix}, \quad \mathbf{B}_0 = 3 \left(\kappa^{\mathrm{in}} - \kappa^{\mathrm{ex}} \right) R_0 \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix}, \quad \mathbf{B}_2 = -2R_0 \left(\mu^{\mathrm{ex}} - \mu^{\mathrm{in}} \right) \begin{bmatrix} 1 & 0 \\ 0 & 6 \end{bmatrix}$$
(3.24)

Finally, using the capillary surface elasticity model, by (2.24) and (3.4) we have Liu et al. (2017):

$$E_{s}[\mathbf{u}] = \sum_{l=0}^{\infty} \frac{1}{2} \begin{bmatrix} X_{l} \\ \Theta_{l} \end{bmatrix} \cdot \mathbf{S}_{l} \begin{bmatrix} X_{l} \\ \Theta_{l} \end{bmatrix}, \qquad \mathbf{S}_{l} = \gamma \begin{bmatrix} l(l+1)-2 & 0 \\ 0 & 0 \end{bmatrix}.$$
(3.25)

Collecting all terms in (3.13), by (3.15)–(3.25) we write the total energy as a quadratic function of (X'_{l}, Θ'_{l}) :

$$F_{1}[\mathbf{u}] = \frac{1}{2} \Big[\nu_{f} \bar{\mathbf{E}} \cdot \mathbb{C}^{\text{in}} \bar{\mathbf{E}} + (1 - \nu_{f}) \bar{\mathbf{E}} \cdot \mathbb{C}^{\text{ex}} \bar{\mathbf{E}} \Big] + \sum_{l=0,2} \frac{1}{2} \begin{bmatrix} \bar{X}_{l} \\ \bar{\Theta}_{l} \end{bmatrix} \cdot \mathbf{S}_{l} \begin{bmatrix} \bar{X}_{l} \\ \bar{\Theta}_{l} \end{bmatrix} + Q_{1} (X_{l}^{\prime}, \Theta_{l}^{\prime}),$$
(3.26)

where

$$Q_1(X_l',\Theta_l') = \sum_{l=0,2} \left\{ \frac{1}{2} \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix} \cdot (\mathbf{D}_l^{\text{in}} + \mathbf{D}_l^{\text{ex}} + \mathbf{S}_l) \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix} + \begin{bmatrix} X_l' \\ \Theta_l' \end{bmatrix} \cdot (\mathbf{S}_l + \mathbf{B}_l) \begin{bmatrix} \bar{X}_l \\ \bar{\Theta}_l \end{bmatrix} \right\}.$$
(3.27)

According to the original variational principle (3.7), the equilibrium X'_0, X'_2, Θ'_2 can be determined by minimizing Q_1 :

$$Q_1^*(a,b) := \min_{X_0', X_2', \Theta_2'} Q_1(X_0', X_2', \Theta_2').$$
(3.28)

Being a quadratic minimization problem, we immediately find the minimizing (X'_0, X'_2, Θ'_2) :

$$X_{0}^{\prime} = \frac{2(2a+b)\sqrt{\pi}R_{0}\left(\frac{2\gamma}{R_{0}} + 3(\kappa^{ex} - \kappa^{in})\right)}{3\left(-\frac{2\gamma}{R_{0}} + 3\kappa^{in} + \frac{3\nu_{f}\kappa^{ex} + 4\mu^{ex}}{1 - \nu_{f}}\right)}, \begin{bmatrix} X_{2}^{\prime} \\ \Theta_{2}^{\prime} \end{bmatrix} = -(\mathbf{D}_{2}^{in} + \mathbf{D}_{2}^{ex} + \mathbf{S}_{2})^{-1}(\mathbf{S}_{2} + \mathbf{B}_{2})\begin{bmatrix} \bar{X}_{2} \\ \bar{\Theta}_{2} \end{bmatrix}.$$
(3.29)

Inserting the solution (3.29) back into (3.27), we find the minimum free energy as a function of (a, b):

$$F_1^*(a,b) = \frac{1}{2}\mathbf{\tilde{E}} \cdot \left[(1-v_f)\mathbb{C}^{\text{ex}} + v_f\mathbb{C}^{\text{in}} \right] \mathbf{\tilde{E}} + \frac{1}{2}\gamma \left(-2\bar{X}_0^2 + 4\bar{X}_2^2 \right) + Q_1^*(a,b)$$

where

$$Q_{1}^{*}(a,b) = -\frac{\bar{X}_{0}^{2}R_{0}(\frac{2\gamma}{R_{0}} + 3(\kappa^{\text{ex}} - \kappa^{\text{in}}))^{2}}{2\left(-\frac{2\gamma}{R_{0}} + 3\kappa^{\text{in}} + \frac{3\nu_{f}\kappa^{\text{ex}} + 4\mu^{\text{ex}}}{1 - \nu_{f}}\right)} - \frac{1}{2}\left[\bar{\mathfrak{Q}}_{2}\right] \cdot (\mathbf{S}_{2} + \mathbf{B}_{2})(\mathbf{D}_{2}^{\text{in}} + \mathbf{D}_{2}^{\text{ex}} + \mathbf{S}_{2})^{-1}(\mathbf{S}_{2} + \mathbf{B}_{2})\left[\bar{\mathfrak{Q}}_{2}\right].$$

It is clear that $Q_1^*(a, b)$ and $F_1^*(a, b)$ are quadratic forms of (a, b). By (3.7) and (3.9), we identify the effective bulk modulus κ^{eff} and effective shear modulus μ^{eff} associated with the effective stiffness tensor \mathbb{C}^{eff} as follows:

$$\kappa^{\text{eff}} = \kappa^{\text{ex}} - \frac{\nu_{\text{f}} \left(\frac{2\gamma}{R_0} + 3(\kappa^{\text{ex}} - \kappa^{\text{in}})\right) (3\kappa^{\text{ex}} + 4\mu^{\text{ex}})}{\frac{6(-1+\nu_{\text{f}})\gamma}{R_0} + 3(3\nu_{\text{f}}(\kappa^{\text{ex}} - \kappa^{\text{in}}) + 3\kappa^{\text{in}} + 4\mu^{\text{ex}})}$$

$$\mu^{\text{eff}} = \mu^{\text{ex}} + 5v_{f}\mu^{\text{ex}}(3\kappa^{\text{ex}} + 4\mu^{\text{ex}}) \left(I_{2}(-\mu^{\text{ex}} + \mu^{\text{in}}) + \frac{2\gamma(I_{1} + 2\mu^{\text{ex}})(6\kappa^{\text{in}} + 17\mu^{\text{in}})}{R_{0}} \right) / (I_{2}(\mu^{\text{ex}}(9 + 6v_{f})\kappa^{\text{ex}} + 4(2 + 3v_{f})\mu^{\text{ex}}) - 6(-1 + v_{f})(\kappa^{\text{ex}} + 2\mu^{\text{ex}})\mu^{\text{in}}) + \frac{12\gamma}{R_{0}}(-6\kappa^{\text{in}}\mu^{\text{ex}}((-5 + 2v_{f})\kappa^{\text{ex}} + 4(-2 + v_{f})\mu^{\text{ex}}) + (-30(-1 + v_{f})\kappa^{\text{ex}}\kappa^{\text{in}} + (85 - 34v_{f})\kappa^{\text{ex}}\mu^{\text{ex}}) - 60(-1 + v_{f})\kappa^{\text{in}}\mu^{\text{ex}} - 68(-2 + v_{f})(\mu^{\text{ex}})^{2})\mu^{\text{in}} - 22(-1 + v_{f})(\kappa^{\text{ex}} + 2\mu^{\text{ex}})(\mu^{\text{in}})^{2})),$$
(3.30)

where, to avoid the bulkiness of the final expressions, we have introduced the constants I_1 and I_2 :

$$I_{1} = \frac{2\mu^{\text{in}}(15\kappa^{\text{in}}+11\mu^{\text{in}})}{6\kappa^{\text{in}}+17\mu^{\text{in}}},$$

$$I_{2} = \kappa^{\text{in}}(48\mu^{\text{ex}}+57\mu^{\text{in}})+4\mu^{\text{in}}(34\mu^{\text{ex}}+\mu^{\text{in}}).$$
(3.31)

We remark that the above formula, though appear to be general, are only applicable to *incompressible* interior with $\kappa^{\text{in}} \rightarrow +\infty$. The above results contain several limiting cases:

1. Dilute volume fraction of incompressible particles:

$$\kappa^{\text{eff}} = \kappa^{\text{ex}} + \frac{\nu_{\text{f}}}{3} (3\kappa^{\text{ex}} + 4\mu^{\text{ex}}),$$

$$\mu^{\text{eff}} = \mu^{\text{ex}} + 5\nu_{\text{f}}\mu^{\text{ex}} (3\kappa^{\text{ex}} + 4\mu^{\text{ex}}) ((48\mu^{\text{ex}} + 57\mu^{\text{in}})(-\mu^{\text{ex}} + \mu^{\text{in}}) + \frac{12\gamma}{R_{0}} (5\mu^{\text{in}} + 2\mu^{\text{ex}})) / ((48\mu^{\text{ex}} + 57\mu^{\text{in}})(\mu^{\text{ex}}(9\kappa^{\text{ex}} + 8\mu^{\text{ex}}) + 6(\kappa^{\text{ex}} + 2\mu^{\text{ex}})\mu^{\text{in}}) + \frac{12\gamma}{R_{0}} (30\kappa^{\text{ex}}(\mu^{\text{ex}} + \mu^{\text{in}}) + 12\mu^{\text{ex}}(4\mu^{\text{ex}} + 5\mu^{\text{in}}))).$$

$$(3.32)$$

2. Rigid particles: The fillers are rigid particles meaning that they approach the following elastic properties $\mu^{in}, \kappa^{in} \to +\infty$

$$\kappa^{\text{eff}} = \kappa^{\text{ex}} + \frac{\nu_{\text{f}}(3\kappa^{\text{ex}} + 4\mu^{\text{ex}})}{3(1 - \nu_{\text{f}})}, \ \mu^{\text{eff}} = \mu^{\text{ex}} + \frac{5\nu_{\text{f}}\mu^{\text{ex}}(3\kappa^{\text{ex}} + 4\mu^{\text{ex}})}{6(1 - \nu_{\text{f}})(\kappa^{\text{ex}} + 2\mu^{\text{ex}})}.$$
(3.33)

In addition, if the matrix is incompressible ($\kappa^{ex} \to +\infty$), then the above equations reduce to $\kappa^{eff} = +\infty$ and

$$\mu^{\text{eff}} = \mu^{\text{ex}} + \frac{5v_{\text{f}}\mu^{\text{ex}}}{2(1 - v_{\text{f}})}.$$
(3.34)

3. Liquid-like particles: In the limiting case of having liquid like particles, the elastic properties of the inclusion will verify $\mu^{in} \rightarrow 0, \kappa^{in} \rightarrow +\infty$

$$\kappa^{\text{eff}} = \kappa^{\text{ex}} + \frac{\nu_{\text{f}}(3\kappa^{\text{ex}} + 4\mu^{\text{ex}})}{3(1 - \nu_{\text{f}})},$$

$$\mu^{\text{eff}} = \mu^{\text{ex}} - \frac{5\nu_{\text{f}}\mu^{\text{ex}} \left(-\frac{\gamma}{R_{0}} + 2\mu^{\text{ex}}\right)(3\kappa^{\text{ex}} + 4\mu^{\text{ex}})}{\frac{3\gamma}{R_{0}}((5 - 2\nu_{\text{f}})\kappa^{\text{ex}} - 4(-2 + \nu_{\text{f}})\mu^{\text{ex}}) + 2\mu^{\text{ex}}((9 + 6\nu_{\text{f}})\kappa^{\text{ex}} + 4(2 + 3\nu_{\text{f}})\mu^{\text{ex}})}.$$
(3.35)

In addition, if the matrix is incompressible ($\kappa^{\text{ex}} \to +\infty$), then the above equations reduce to $\kappa^{\text{eff}} = +\infty$, and

$$\mu^{\text{eff}} = \mu^{\text{ex}} - \frac{5v_{\text{f}}\mu^{\text{ex}} \left(-\frac{\gamma}{R_{0}} + 2\mu^{\text{ex}}\right)}{\frac{\gamma}{R_{0}} (5 - 2v_{\text{f}}) + 2(3 + 2v_{\text{f}})\mu^{\text{ex}}}.$$
(3.36)

3.4. Results and discussion

The first observation that we can make is that the capillary effect does not manifest if we are dealing with rigid particles. In contrast, the capillary effect is very important when the inclusions behave like liquid-like particles embedded in a compliant matrix, see Fig. 5. In the dilute limit, our results, in particular the effective Young's modulus shown below, match those obtained by Style et al. (2015b):

$$\frac{E^{\text{eff}}}{E^{\text{ex}}} = 1 + \frac{5\nu_{\text{f}}\left(\frac{3\gamma}{E^{\text{ex}}R_{0}} - 2\right)}{3\left(\frac{5\gamma}{E^{\text{ex}}R_{0}} + 2\right)}.$$
(3.37)

As well-evident in Fig. 5, the conventional wisdom that would suggest the softening of a compliant matrix filled with liquid inclusions is only true for low values of the dimensionless capillary number $\gamma/(E^{ex}R_0)$ —i.e. when either the inclusions are large or surface tension is small. In fact, as already noted by Style et al. (2015b), when the surface tension is negligible, the composite (in the dilute limit) become more compliant in exact agreement with Eshelby's prediction of $E^{\text{eff}}/E^{\text{ex}} = (1 - 5/3v_f)$ (Eshelby, 1957). On the other hand, in the surface tension dominated limit, $E^{\text{eff}}/E^{\text{ex}} = (1 + v_f)$ in accordance with Style et al. (2015b). The overall stiffening of the composite due to capillarity could be understood due to the geometry of the inclusions. Starting from a spherical shape, the surface tension tends to keep this preferred geometry to ensure the minimum energy. However, any tension or compression applied to the composite will try to deform the liquid-like spheres



Fig. 5. Effective properties of an incompressible matrix filled with liquid-like spherical particles ($\kappa^{\text{ex}} = +\infty, \mu^{\text{in}} = 0, \kappa^{\text{in}} = +\infty$). The left plot shows the variation of the effective shear modulus with respect to the volume fraction while the one on the right depicts the result for the effective Young's modulus in the dilute limit as shown in Eq. (3.37).

to ellipsoids and take them out from their minimum energy configuration. In this sense, the surface tension will resist the deformation which will result in an overall stiffening of the composite.

We close this section by making a few general remarks on the significance of our formulation and subsequent linearization of the capillary theory for soft materials. First and foremost our formulation is consistent from a classical continuum mechanics viewpoint and it becomes quite clear *how* and (more importantly) *why* residual surface tension can impact effective mechanical properties. Finally, the construction of the presented framework allows for a systematic extension to the electrostatic case—which we present next.

4. Effective electrostrictive properties of composites with capillary effects

4.1. Homogenization formulation and effective electrostrictive properties

We now consider the effective electro-elastic properties of the soft composite. In the absence of capillary effects, the homogenization and effective electrostrictive properties have been addressed in Lefevre and Lopez-Pamies (2014) and Tian (2007). Two of the authors, Liu and Sharma (2018) have recently explored the emergent piezoelectric properties for composites with embedded charges and dipoles. In this work, we focus on the effective electrostrictive properties taking into account of the capillary effects from interfaces between different phases. The motivation lies in the physical intuition that soft and highly polarizable materials permit large electrostrictive coupling coefficients. Liquid ionic inclusions in soft matter are clearly the extremes of soft and highly polarizable materials. Evidently, as discussed in the last section, the interfaces between liquid inclusions and ambient soft matrix may contribute non-negligible elastic energy and significantly influence the overall electrostrictive properties of the composite, necessitating the subsequent calculation of effective electrostrictive electrostrictive properties with capillary effects.

As in the preceding section, we employ a variational formulation and the principle of energy equivalence to define the effective properties of the composite. Let ϵ^{ex} (resp. ϵ^{in}) be the permittivity of the matrix (resp. inclusion) phase and define the **x**-dependent permittivity $\epsilon_{\#}: Y \to \mathbb{R}$ as

$$\epsilon_{\#} = \begin{cases} \epsilon^{\mathrm{ex}} & \mathrm{in} \ Y \setminus \Omega_0 \\ \epsilon^{\mathrm{in}} & \mathrm{in} \ \Omega_0. \end{cases}$$

If the average electric field in the RVE is prescribed at $\mathbf{\bar{e}} \in \mathbb{R}$, the local electric field $-\nabla \xi_{\mathbf{\bar{e}}} : Y \to \mathbb{R}$ is uniquely determined by the variational problem

$$G^{\text{eff}}(\tilde{\mathbf{e}}) := \min_{\boldsymbol{\xi} \in \mathcal{P}_{\tilde{\mathbf{e}}}} \left\{ G[\boldsymbol{\xi}] = \int_{Y} \frac{\epsilon_{\#}}{2} |\nabla \boldsymbol{\xi}|^{2} \right\},\tag{4.1}$$

where the admissible space $\mathcal{P}_{\tilde{e}}$ for potential is defined as

$$\mathcal{U}_{\bar{\mathbf{e}}} = \{ \xi : -\int_{Y} \nabla \xi = \bar{\mathbf{e}}, \qquad \nabla \xi \text{ is } Y \text{-periodic} \}.$$
(4.2)

Let $\xi_{\tilde{\mathbf{e}}} \in \mathcal{P}_{\tilde{\mathbf{e}}}$ be a minimizer to (4.1). Clearly, it should satisfy the associated Euler–Lagrange equation:

$$\nabla \cdot (\epsilon_{\#} \nabla \xi_{\bar{\mathbf{e}}}) = 0 \qquad \text{in } Y.$$
(4.3)

Also, the effective permittivity tensor ϵ^{eff} of the composite is related with the minimum energy $G^{\text{eff}}(\bar{\mathbf{e}})$ by

$$G^{\text{eff}}(\mathbf{\bar{e}}) = \frac{1}{2} \mathbf{\bar{e}} \cdot \boldsymbol{\epsilon}^{\text{eff}} \mathbf{\bar{e}}, \qquad \text{i.e.,} \qquad \boldsymbol{\epsilon}^{\text{eff}} = \frac{\partial^2 G^{\text{eff}}(\mathbf{\bar{e}})}{\partial \mathbf{\bar{e}} \partial \mathbf{\bar{e}}}.$$
(4.4)

For ideal dielectric materials, the electrostrictive stress due to the electric field can be written as Liu (2014), Liu and Sharma (2018), Tian (2007), and Tian et al. (2012):

$$\boldsymbol{\sigma}_{\text{elect}} = \mathbb{A}_{\#}(\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}), \tag{4.5}$$

where the fourth-order tensor $A_{\#}$ is given by

$$(\mathbb{A}_{\#})_{ijkl} = \frac{\epsilon_{\#}}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \delta_{ij} \delta_{kl}), \quad \text{i.e.,} \quad \mathbb{A}_{\#} (\nabla \xi \otimes \nabla \xi) = \epsilon_{\#} \nabla \xi \otimes \nabla \xi - \frac{\epsilon_{\#}}{2} |\nabla \xi|^{2} \mathbf{I}.$$
(4.6)

By (4.3), we immediately find that

$$\operatorname{div}\boldsymbol{\sigma}_{\operatorname{elect}} = 0 \qquad \text{in } Y \setminus (\partial \Omega_0). \tag{4.7}$$

In other words, the electrostrictive stress is equivalent to an interfacial traction on $\partial \Omega_0$.

If the average strain in the RVE is prescribed at $\mathbf{\bar{E}} \in \mathbb{R}^{3 \times 3}_{sym}$, the actual local strain is uniquely determined by the following variational¹⁹ problem (Tian, 2007; Tian et al., 2012):

$$\Psi_2^{\text{eff}}(\bar{\mathbf{E}}, \bar{\mathbf{e}}) := \min\{F_2[\mathbf{u}; \bar{\mathbf{e}}]: \mathbf{u} \in \mathcal{U}_{\bar{\mathbf{E}}}\},\tag{4.8}$$

where the energy $F_2[\mathbf{u}; \mathbf{\bar{e}}]$ is given by

$$F_{2}[\mathbf{u};\bar{\mathbf{e}}] = E_{s}[\mathbf{u}] + \int_{Y} \frac{1}{2} \nabla \mathbf{u} \cdot \mathbb{C}_{\#} \nabla \mathbf{u} d\mathbf{x} + \int_{Y} \nabla \mathbf{u} \cdot \boldsymbol{\sigma}_{\text{elect}} d\mathbf{x}.$$
(4.9)

We remark that in the above equation, the first term arises from the capillary model for the interface $\partial \Omega_0$ (cf., (3.4)), the second term is the bulk elastic energy, and the last term reflects the electrostrictive effect of the electric field.

We are interested in the effective electrostrictive properties of the composite and how surface tension γ influences the macroscopic electrostriction. Following the classic paradigm of homogenization, we define the effective stiffness tensor \mathbb{C}^{eff} and the effective electrostrictive tensor \mathbb{A}^{eff} by equating the energy density of the composite to the energy density of "homogenized" bulk material with the same average strain and electric field:

$$\Psi_{2}^{\text{eff}}(\mathbf{\tilde{E}}, \mathbf{\tilde{e}}) = \Psi_{2}^{\text{eff}}(0, 0) + \boldsymbol{\sigma}^{0} \cdot \mathbf{\tilde{E}} + \frac{1}{2} \mathbf{\tilde{E}} \cdot \mathbb{C}^{\text{eff}} \mathbf{\tilde{E}} + \mathbf{\tilde{E}} \cdot \mathbb{A}^{\text{eff}}(\mathbf{\tilde{e}} \otimes \mathbf{\tilde{e}}) + \cdots$$
(4.10)

In other words, we define the effective tensors \mathbb{C}^{eff} , \mathbb{A}^{eff} by

$$\mathbb{C}^{\text{eff}} = \frac{\partial^2 \Psi_2^{\text{eff}}}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{E}}} \Big|_{(\bar{\mathbf{E}}, \bar{\mathbf{e}})=0} \quad \text{and} \quad \mathbb{A}^{\text{eff}} = \frac{1}{2} \frac{\partial^3 \Psi_2^{\text{eff}}}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{e}} \partial \bar{\mathbf{e}}} \Big|_{(\bar{\mathbf{E}}, \bar{\mathbf{e}})=0}. \tag{4.11}$$

Comparing the functional form (4.9) with (3.5), we find that the effective stiffness tensor \mathbb{C}^{eff} is exactly the same as in Section 3, and the effective electrostrictive tensor \mathbb{A}^{eff} can be determined by

$$\mathbb{A}^{\text{eff}} = \frac{1}{2} \left\{ \frac{\partial^3}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{e}} \partial \bar{\mathbf{e}}} F_2[\mathbf{u}_{\bar{\mathbf{E}}}; \bar{\mathbf{e}}] \right\} \Big|_{(\bar{\mathbf{E}}, \bar{\mathbf{e}})=0}, \tag{4.12}$$

where $u_{\tilde{E}} \in \mathcal{U}_{\tilde{E}}$ is a minimizer of (4.8) and satisfies the associated Euler–Lagrange equations:

$$\begin{cases} \operatorname{div}(\mathbb{C}^{\mathrm{ex}}\nabla\mathbf{u} + \boldsymbol{\sigma}_{\mathrm{elect}}) = \operatorname{div}(\mathbb{C}^{\mathrm{ex}}\nabla\mathbf{u}) = 0 & \text{ in } Y \setminus \Omega_0, \\ \operatorname{div}(\mathbb{C}^{\mathrm{in}}\nabla\mathbf{u} + \boldsymbol{\sigma}_{\mathrm{elect}}) = \operatorname{div}(\mathbb{C}^{\mathrm{in}}\nabla\mathbf{u}) = 0 & \text{ in } \Omega_0. \end{cases}$$

$$(4.13)$$

For isotropic composites, the electrostrictive tensor \mathbb{A}^{eff} has to be of the same form as an isotropic elasticity tensor and can be written as

$$\mathbb{A}^{\text{eff}} = m_{K}^{\text{eff}} \mathcal{K} + m_{J}^{\text{eff}} \mathcal{J} \quad \text{where}$$

$$\mathcal{K}_{ijkl} = \frac{1}{2} \Big[\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl} \Big] \quad \text{and} \quad \mathcal{J}_{ijkl} = \frac{1}{3} \delta_{ij} \delta_{kl}$$

$$(4.14)$$

where m_K^{eff} , m_J^{eff} are two material constants in analogy with shear and bulk moduli of an isotropic elastic material. Immediately, we find the relations between the component of tensor \mathbb{A}^{eff} and constants m_K^{eff} , m_J^{eff} satisfy

$$\mathbb{A}_{1133}^{\text{eff}} = -\frac{1}{3}m_{K}^{\text{eff}} + \frac{1}{3}m_{J}^{\text{eff}}, \quad \mathbb{A}_{1111}^{\text{eff}} = \frac{2}{3}m_{K}^{\text{eff}} + \frac{1}{3}m_{J}^{\text{eff}}.$$
(4.15)

¹⁹ The electrostriction homogenization problem is complicated by fact that it is nonlinear in nature. Tian (2007) and Tian et al. (2012) introduced a clever scaling ansatz that allows a decoupling of the mechanical and electrical problems and the variational principle used below is a consequence of that. Due to this scaling assumption, and as evident from the variational principle, the electrical problem is solved separately and its results then enter as a "forcing" function in the mechanical problem.

4.2. Exact solutions for a coated sphere with surface energy

For exact solutions, we consider the special microstructure of Hashin's coated spheres. For a coated sphere as illustrated in Fig. 4(b), the condition $-\int_{Y} \nabla \xi = \bar{\mathbf{e}}$ should be replaced by

$$\int_{\{r=R\}} \xi \mathbf{e}_r = -\frac{4\pi R^3}{3} \bar{\mathbf{e}}.$$
(4.16)

If the average electric field $\mathbf{\bar{e}} = e_0 \mathbf{e}_z$, the solution to the electrostatic problem (4.3) and (4.16) is well-known and can be found in the textbook, e.g., Milton (2002):

$$\xi_{\tilde{\mathbf{e}}} = \begin{cases} -c_0^{\text{in}} e_0 r \cos\theta & \text{if } r \le R_0, \\ -c_1^{\text{ex}} e_0 r \cos\theta - c_2^{\text{ex}} e_0 \frac{\cos\theta}{r^2} & \text{if } R_0 < r < R. \end{cases}$$

$$(4.17)$$

where

$$c_0^{\text{in}} = \frac{3\epsilon^{\text{ex}}}{(\nu_f + 2)\epsilon^{\text{ex}} - \nu_f \epsilon^{\text{in}} + \epsilon^{\text{in}}},$$

$$c_1^{\text{ex}} = \frac{2\epsilon^{\text{ex}} + \epsilon^{\text{in}}}{(\nu_f + 2)\epsilon^{\text{ex}} - \nu_f \epsilon^{\text{in}} + \epsilon^{\text{in}}},$$

$$c_2^{\text{ex}} = \frac{R_0^3(\epsilon^{\text{ex}} - \epsilon^{\text{in}})}{(\nu_f + 2)\epsilon^{\text{ex}} - \nu_f \epsilon^{\text{in}} + \epsilon^{\text{in}}}.$$

Immediately, from the definition (4.4) we find the effective permittivity of the composite is given by

$$\epsilon^{\text{eff}} = \epsilon^{\text{ex}} + \frac{3\nu_{\text{f}}\epsilon^{\text{ex}}(-\epsilon^{\text{ex}} + \epsilon^{\text{in}})}{(2 + \nu_{\text{f}})\epsilon^{\text{ex}} + \epsilon^{\text{in}} - \nu_{\text{f}}\epsilon^{\text{in}}}.$$
(4.18)

This result is classic and matches the Hashin–Shtrikman's bounds for any volume fraction $v_f \in (0, 1)$.

To solve the minimization problem (4.8), it is convenient to rewrite the last term in (4.9) as a surface integral. First, we notice that

$$\int_{Y} \nabla \mathbf{u} \cdot \boldsymbol{\sigma}_{\text{elect}} = \int_{Y} (\mathbf{\tilde{E}} + \nabla \mathbf{u}') \cdot \boldsymbol{\sigma}_{\text{elect}} = \int_{Y} \mathbf{\tilde{E}} \cdot \boldsymbol{\sigma}_{\text{elect}} - \int_{\partial \Omega_{0}} \mathbf{u}' \cdot [\![\boldsymbol{\sigma}_{\text{elect}}]\!] \mathbf{n}.$$
(4.19)

From (3.19) and (A.9), we find that

$$\int_{Y} \tilde{\mathbf{E}} \cdot \boldsymbol{\sigma}_{\text{elect}} = \frac{2}{3} \pi R_{0}^{3}(e_{0})^{2} \epsilon^{\text{in}}(c_{0}^{\text{in}})^{2}(b-2a) + \frac{2\pi e_{0}^{2} R_{0}^{3}(v_{\text{f}}-1)\epsilon^{\text{ex}}}{15v_{\text{f}}((v_{\text{f}}+2)\epsilon^{\text{ex}} - v_{\text{f}}\epsilon^{\text{in}} + \epsilon^{\text{in}})^{2}} (2(\epsilon^{\text{ex}})^{2}(4a(v_{\text{f}}+5) + b(v_{\text{f}}-10)) - 4\epsilon^{\text{ex}}\epsilon^{\text{in}}(2a(2v_{\text{f}}-5) + b(v_{\text{f}}+5)) + (\epsilon^{\text{in}})^{2}(2a(4v_{\text{f}}+5) + b(2v_{\text{f}}-5))).$$

$$(4.20)$$

In addition, by (A.9) and (A.27) we obtain

$$-\int_{\partial\Omega_0} \mathbf{u}' \cdot \left[\!\left[\boldsymbol{\sigma}_{\text{elect}}\right]\!\right] \mathbf{n} = \sum_{l=0,2} \left\{ \mathbf{b}_l \cdot \left[\begin{matrix} X_l' \\ \Theta_l' \end{matrix}\right] \right\},\tag{4.21}$$

where

$$\mathbf{b}_{0} = \left[\frac{3\sqrt{\pi}e_{0}^{2}R_{0}^{2}\epsilon^{\exp}\left(2(\epsilon^{\exp})^{2} - \epsilon^{\exp}\epsilon^{\sin} - (\epsilon^{\sin})^{2}\right)}{((\nu_{f}+2)\epsilon^{\exp} - \nu_{f}\epsilon^{\sin} + \epsilon^{\sin})^{2}}, 0\right], \mathbf{b}_{2} = \left[-\frac{6\sqrt{\frac{\pi}{5}}e_{0}^{2}R_{0}^{2}\epsilon^{\exp}(\epsilon^{\exp} - \epsilon^{\sin})^{2}}{((\nu_{f}+2)\epsilon^{\exp} - \nu_{f}\epsilon^{\sin} + \epsilon^{\sin})^{2}}, 0\right].$$
(4.22)

Collecting terms in (4.9), by (3.15), (3.20)–(3.25), and (4.19)–(4.22), we once again manage to write the total energy as a quadratic function of (X'_l, Θ'_l) :

$$F_{2}[\mathbf{u};\bar{\mathbf{e}}] = \frac{1}{2}\bar{\mathbf{E}} \cdot [\nu_{\rm f}\mathbb{C}^{\rm in} + (1-\nu_{\rm f})\cdot\mathbb{C}^{\rm ex}]\bar{\mathbf{E}} + \frac{1}{2}\gamma(-2\bar{X}_{0}^{2} + 4\bar{X}_{2}^{2}) + \bar{\mathbf{E}}\cdot\int_{Y}\boldsymbol{\sigma}_{\rm elect} + Q_{2}(X_{l}',\Theta_{l}'), \qquad (4.23)$$

where

$$Q_{2}(X_{l}',\Theta_{l}') = Q_{1}(X_{l}',\Theta_{l}') + \sum_{l=0,2} \left\{ \mathbf{b}_{l} \cdot \begin{bmatrix} X_{l}'\\\Theta_{l}' \end{bmatrix} \right\} = \frac{1}{2} \sum_{l=0,2} \left\{ \begin{bmatrix} X_{l}'\\\Theta_{l}' \end{bmatrix} \cdot (\mathbf{D}_{l}^{\text{in}} + \mathbf{D}_{l}^{\text{ex}} + \mathbf{S}_{l}) \begin{bmatrix} X_{l}'\\\Theta_{l}' \end{bmatrix} + \begin{bmatrix} X_{l}'\\\Theta_{l}' \end{bmatrix} \cdot \left\{ (\mathbf{S}_{l} + \mathbf{B}_{l}) \begin{bmatrix} \bar{X}_{l}\\\bar{\Theta}_{l} \end{bmatrix} + \mathbf{b}_{l} \right\} \right\}.$$

$$(4.24)$$

From the original variational principle (4.8), the equilibrium X'_0, X'_2, Θ'_2 can be determined by minimizing Q_2 :

$$Q_2^*(a,b) := \min_{X_0', X_2', \Theta_2'} Q_2(X_0', X_2', \Theta_2').$$
(4.25)

Being a quadratic minimization problem, we immediately find the minimizing (X'_0, X'_2, Θ'_2) :

$$X_{0}' = -\frac{\sqrt{\pi}R_{0}\left(\frac{3}{\sqrt{\pi}R_{0}^{2}}(\mathbf{b}_{0})_{1} - \frac{4b\gamma}{R_{0}} - 4a\left(\frac{2\gamma}{R_{0}} + 3(\kappa^{ex} - \kappa^{in})\right) + 6b(-\kappa^{ex} + \kappa^{in})\right)}{3\left(-\frac{2\gamma}{R_{0}} + 3\kappa^{in} - \frac{3v_{f}\kappa^{ex} + 4\mu^{ex}}{-1 + v_{f}}\right)},$$

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$$\begin{bmatrix} X_2'\\ \Theta_2' \end{bmatrix} = -(\mathbf{D}_2^{\text{in}} + \mathbf{D}_2^{\text{ex}} + \mathbf{S}_2)^{-1} \left\{ (\mathbf{S}_2 + \mathbf{B}_2) \begin{bmatrix} \bar{X}_2\\ \bar{\Theta}_2 \end{bmatrix} + \mathbf{b}_2 \right\},\tag{4.26}$$

where $(\mathbf{b}_0)_1$ refers to first component of \mathbf{b}_0 (cf., (4.22)). Inserting the solution (4.26) back into (4.25), we find the minimum free energy as a function of (a, b):

$$F_{2}^{*}(a,b) = \frac{1}{2}\bar{\mathbf{E}} \cdot [(1-\nu_{\rm f})\mathbb{C}^{\rm ex} + \nu_{\rm f}\mathbb{C}^{\rm in}]\bar{\mathbf{E}} + \frac{1}{2}\gamma(-2\bar{X}_{0}^{2} + 4\bar{X}_{2}^{2}) + \bar{\mathbf{E}} \cdot \int_{Y} \boldsymbol{\sigma}_{\rm elect} + Q_{2}^{*}(a,b),$$

where

$$\begin{aligned} Q_2^*(a,b) &= Q_1^*(a,b) - \frac{1}{2} \frac{(\mathbf{b}_0)_1}{(\mathbf{D}_0^{\text{in}})_{11} + (\mathbf{D}_0^{\text{ex}})_{11} - 2\gamma} \Big[2(-2\gamma + (\mathbf{B}_0)_{11}) \bar{X}_0 + (\mathbf{b}_0)_1 \Big] \\ &- \frac{1}{2} \mathbf{b}_2 \cdot (\mathbf{D}_2^{\text{in}} + \mathbf{D}_2^{\text{ex}} + \mathbf{S}_2)^{-1} \bigg[2(\mathbf{S}_2 + \mathbf{B}_2) \bigg[\frac{\bar{X}_2}{\bar{\Theta}_2} \bigg] + \mathbf{b}_2 \bigg]. \end{aligned}$$

We remark that $Q_2^*(a, b)$ and $F_2^*(a, b)$ are not quadratic functions of e_0 . Nevertheless, from (4.10) and (4.11) we can identify the effective electrostrictive tensor \mathbb{A}^{eff} by

$$\bar{\mathbf{E}} \cdot \mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) = \bar{\mathbf{E}} \cdot \int_{Y} \boldsymbol{\sigma}_{\text{elect}} - \sum_{l=0,2} \left\{ \mathbf{b}_{l} \cdot (\mathbf{D}_{l}^{\text{in}} + \mathbf{D}_{l}^{\text{ex}} + \mathbf{S}_{l})^{-1} (\mathbf{S}_{l} + \mathbf{B}_{l}) \begin{bmatrix} \bar{X}_{l} \\ \bar{\Theta}_{l} \end{bmatrix},$$
(4.27)

and in particular, explicitly calculate the components $\mathbb{A}_{1133}^{\text{eff}}$ and $\mathbb{A}_{3333}^{\text{eff}}$. By (4.15), we can then find m_K^{eff} , m_J^{eff} as follows (see details in Appendix A.3):

$$m_{K}^{\text{eff}} = \epsilon^{\text{ex}} + \Delta \epsilon^{2} \frac{1}{15 \nu_{f}} \left(\frac{5 - 2\nu_{f}}{\epsilon^{\text{ex}}} + \frac{15}{\epsilon^{\text{in}} - \epsilon^{\text{ex}}} \right) + \Delta \epsilon^{2} \Delta \mu \left(-\frac{1}{5 \nu_{f}^{2} \epsilon^{\text{ex}} \left(-\mu^{\text{ex}} + \mu^{\text{in}} + \frac{4\gamma}{R_{0}} \frac{6\kappa^{\text{in}}\mu^{\text{ex}} + 15\kappa^{\text{in}}\mu^{\text{in}} + 17\mu^{\text{ex}}\mu^{\text{in}} + 11(\mu^{\text{in}})^{2}}{I_{2}} \right)}{5 \nu_{f}^{2} \epsilon^{\text{ex}} \left(-\mu^{\text{ex}} + \mu^{\text{in}} + \frac{4\gamma}{R_{0}} \frac{6\kappa^{\text{in}}\mu^{\text{ex}} + 15\kappa^{\text{in}}\mu^{\text{in}} + 17\mu^{\text{ex}}\mu^{\text{in}} + 11(\mu^{\text{in}})^{2}}{I_{2}} \right)} \right)$$
$$m_{J}^{\text{eff}} = -\frac{\epsilon^{\text{ex}}}{2} - \frac{1}{2} \Delta \epsilon - \Delta \epsilon^{2} \frac{(-1 + \nu_{f}) \left(2\epsilon^{\text{ex}} + \epsilon^{\text{in}}\right) \left(\frac{2\gamma}{R_{0}} + 3\left(\kappa^{\text{ex}} - \kappa^{\text{in}}\right)\right)}{2\nu_{f} \epsilon^{\text{ex}} \left(\epsilon^{\text{ex}} - \epsilon^{\text{in}}\right) \left(\frac{2(-1 + \nu_{f})\gamma}{R_{0}} + 3\nu_{f} \left(\kappa^{\text{ex}} - \kappa^{\text{in}}\right) + 3\kappa^{\text{in}} + 4\mu^{\text{ex}}} \right)},$$
(4.28)

where I_2 is defined in (3.31) and with $\Delta \epsilon = \epsilon^{\text{eff}} - \epsilon^{\text{ex}}$ and $\Delta \mu = \mu^{\text{eff}} - \mu^{\text{ex}}$.

The above results contain several limiting cases:

1. Dilute volume fraction of incompressible particles:

$$\begin{split} m_{K}^{\text{eff}} &= \epsilon^{\text{ex}} + \frac{3v_{\text{f}}\epsilon^{\text{ex}}(-\epsilon^{\text{ex}} + \epsilon^{\text{in}})}{2\epsilon^{\text{ex}} + \epsilon^{\text{in}}} - \left(9v_{\text{f}}\epsilon^{\text{ex}}(\epsilon^{\text{ex}} - \epsilon^{\text{in}})^{2}\mu^{\text{ex}}(3\kappa^{\text{ex}} + 4\mu^{\text{ex}})\right) \Big/ \\ (2\epsilon^{\text{ex}} + \epsilon^{\text{in}})^{2} \left(\mu^{\text{ex}}(9\kappa^{\text{ex}} + 8\mu^{\text{ex}}) + 6(\kappa^{\text{ex}} + 2\mu^{\text{ex}})\mu^{\text{in}} + \frac{12\gamma}{R_{0}(48\mu^{\text{ex}} + 57\mu^{\text{in}})} \left(30\kappa^{\text{ex}}(\mu^{\text{ex}} + \mu^{\text{in}}) + 12\mu^{\text{ex}}(4\mu^{\text{ex}} + 5\mu^{\text{in}})\right)\right), \\ m_{J}^{\text{eff}} &= \frac{\epsilon^{\text{ex}}}{2} \left(-1 - \frac{6v_{f}(\epsilon^{\text{ex}} - \epsilon^{\text{in}})}{(2\epsilon^{\text{ex}} + \epsilon^{\text{in}})}\right). \end{split}$$
(4.29)

2. Rigid particles with infinite permittivity: In the limit of rigid infinite-permittivity particles when $\mu^{in}, \kappa^{in} \to +\infty$ and $\epsilon^{in} \to +\infty$

$$m_{K}^{\text{eff}} = \epsilon^{\text{ex}} \left(1 + \frac{3(5 - 2\nu_{f})\nu_{f}}{5(1 - \nu_{f})^{2}} \right), \\ m_{J}^{\text{eff}} = \frac{\epsilon^{\text{ex}}}{2} \left(-1 + \frac{3\nu_{f}(2 + \nu_{f})}{(1 - \nu_{f})^{2}} \right).$$
(4.30)

3. Liquid-like particles with infinite permittivity, i.e., $\mu^{in} \rightarrow 0$, $\kappa^{in} \rightarrow +\infty$ and $\epsilon^{in} \rightarrow +\infty$:

$$m_{\kappa}^{\text{eff}} = \epsilon^{\text{ex}} + \frac{3v_{f}\epsilon^{\text{ex}}\left(-\frac{3y}{R_{0}}\left(-5+2v_{f}\right)\left((-5+2v_{f})\kappa^{\text{ex}}+4\left(-2+v_{f}\right)\mu^{\text{ex}}\right)-8\left(1-v_{f}\right)\mu^{\text{ex}}\left(-5\mu^{\text{ex}}+3v_{f}\left(\kappa^{\text{ex}}+2\mu^{\text{ex}}\right)\right)\right)}{5\left(-1+v_{f}\right)^{2}\left(\frac{3y}{R_{0}}\left((-5+2v_{f})\kappa^{\text{ex}}+4\left(-2+v_{f}\right)\mu^{\text{ex}}\right)-2\mu^{\text{ex}}\left((9+6v_{f})\kappa^{\text{ex}}+4\left(2+3v_{f}\right)\mu^{\text{ex}}\right)\right)},$$

$$m_{J}^{\text{eff}} = -\frac{\epsilon^{\text{ex}}}{2} + \frac{3v_{f}(2+v_{f})\epsilon^{\text{ex}}}{2\left(1-v_{f}\right)^{2}}.$$
 (4.31)

In addition, if the matrix is incompressible ($\kappa^{ex} \rightarrow +\infty$), then the above equations reduce to

$$m_{K}^{\text{eff}} = \epsilon^{\text{ex}} - \frac{3\nu_{f}\epsilon^{\text{ex}}\left(\frac{\gamma}{R_{0}}(5-2\nu_{f})^{2} - 8(-1+\nu_{f})\nu_{f}\mu^{\text{ex}}\right)}{5(-1+\nu_{f})^{2}\left(\frac{\gamma}{R_{0}}(-5+2\nu_{f}) - 2(3+2\nu_{f})\mu^{\text{ex}}\right)}, \\ m_{J}^{\text{eff}} = -\frac{\epsilon^{\text{ex}}}{2} + \frac{3\nu_{f}(2+\nu_{f})\epsilon^{\text{ex}}}{2(1-\nu_{f})^{2}}.$$
(4.32)

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Fig. 6. Electrostriction of a film under the application of an electric field.

4.3. Results and discussion

To physically interpret the derived effective electrostriction coefficients, we consider a dielectric thin film as sketched in Fig. 6. The thin film is placed between two compliant electrodes and a uniform external electric field $\mathbf{\tilde{e}} \approx \frac{\phi}{L_3} \mathbf{k}$ is applied, where ϕ denotes the voltage applied between the compliant electrodes and L_3 represents the initial thickness of the thin layer of the dielectric elastomer composite. From (4.13)_{1,2}, the total stress in the film can be expressed as

$$\boldsymbol{\sigma} = \mathbb{C}^{\text{eff}} \tilde{\mathbf{E}} + \mathbb{A}^{\text{eff}} (\tilde{\mathbf{e}} \otimes \tilde{\mathbf{e}}). \tag{4.33}$$

Since there is no applied boundary traction, in equilibrium we necessarily have $\sigma = 0$ which, by (4.33), implies that the average strain $\tilde{\mathbf{E}}$ can be written as

$$\bar{\mathbf{E}} = -\frac{m_{K}^{\text{eff}}}{2\mu^{\text{eff}}} \bar{\mathbf{e}} \otimes \bar{\mathbf{e}} + \left(\frac{m_{K}^{\text{eff}}}{6\mu^{\text{eff}}} - \frac{m_{J}^{\text{eff}}}{9\kappa^{\text{eff}}}\right) (\bar{\mathbf{e}} \cdot \bar{\mathbf{e}}) \mathbf{I}.$$
(4.34)

To be more specific, we can rewrite the strain tensor with respect to the cartesian basis vector $\{i, j, k\}$ as

$$\bar{\mathbf{E}} = \bar{E}_{11}\mathbf{i} \otimes \mathbf{i} + \bar{E}_{22}\mathbf{j} \otimes \mathbf{j} + \bar{E}_{33}\mathbf{k} \otimes \mathbf{k}, \tag{4.35}$$

where

$$\bar{E}_{11} = \bar{E}_{22} = \left(\frac{m_K^{\text{eff}}}{6\mu^{\text{eff}}} - \frac{m_J^{\text{eff}}}{9\kappa^{\text{eff}}}\right) e_0^2, \qquad \bar{E}_{33} = -\left(\frac{m_K^{\text{eff}}}{3\mu^{\text{eff}}} + \frac{m_J^{\text{eff}}}{9\kappa^{\text{eff}}}\right) e_0^2, \tag{4.36}$$

which may be compared with the strains we would obtain if the film were made of the homogeneous soft matrix material:

$$E_{11}^{\text{ex}} = E_{22}^{\text{ex}} = \left(\frac{\epsilon^{\text{ex}}}{6\mu^{\text{ex}}} + \frac{\epsilon^{\text{ex}}}{18\kappa^{\text{ex}}}\right)e_0^2, \qquad E_{33}^{\text{ex}} = -\left(\frac{\epsilon^{\text{ex}}}{3\mu^{\text{ex}}} - \frac{\epsilon^{\text{ex}}}{18\kappa^{\text{ex}}}\right)e_0^2. \tag{4.37}$$

The ratios $\frac{\tilde{E}_{11}}{\frac{E_{33}}{\Gamma_{11}}}$ and $\frac{\tilde{E}_{33}}{\frac{E_{33}}{\Gamma_{33}}}$ thus provide a measure of the electrostrictive deformation due to the presence of capillary effect and liquid inclusions.

4.4. Results for the capillarity and electrostrictive response of a soft matrix containing high permittivity liquid inclusions

We first consider the case of a nearly incompressible dielectric elastomer filled with liquid-like high permittivity spherical particles such that $\kappa^{\text{ex}}/\mu^{\text{ex}} = 10^3$, $\mu^{\text{in}}/\mu^{\text{ex}} = 10^{-2}$, $\kappa^{\text{in}}/\mu^{\text{ex}} = 10^3$, $\epsilon^{\text{in}}/\epsilon^{\text{ex}} = 10^2$ as shown in Fig. 7. These values are within the specific range of dielectric elastomer composites filled with incompressible fluid fillers with vanishingly shear modulus. The behavior of Young's modulus is similar to what was earlier depicted in Fig. 5, since the effective mechanical properties are not affected by electrostriction. According, we focus on analyzing the effect of capillarity on the electrostrictive properties represented in (4.28). As evident from Fig. 7, both electrostriction coefficients m_{K}^{eff} and m_{j}^{eff} increase with increasing value of the capillary number $\gamma/(\mu^{\text{ex}}R_0)$ which would indicate an increase in the propensity of the composite to deform under the action of an electrical field. *However*, the overall stiffening of the composite tends to counteract the deformation. These mutually contradictory effects lead to the situation that the ratio $\bar{E}_{33}/E_{33}^{\text{ex}}$ is relatively uninfluenced by capillary effects. In short, while adding liquid inclusions to a soft solid does lead to enhanced electromechanical coupling, deformation and electrostriction, the effect of *capillarity* is relatively small due to two mutually counteracting effects. This begs the question whether capillarity has an effect on the electromechanical coupling of any physical consequence. We find an affirmative answer to this question in the context of maximum energy of conversion discussion in the next section.

4.4.1. Maximum energy conversion

While capillarity does not appear to influence the actuator action of the soft composite with liquid inclusion, since the electrostriction coefficient does appear to be strongly enhanced, we may consider whether augmented energy conversion is possible. Unlike piezoelectrics, special conditions must be used for energy harvesting through electrostriction. For example,



Fig. 7. Effective properties of a nearly incompressible dielectric elastomer filled with liquid-like high permittivity spherical particles. ($\kappa^{ex} = 10^3 \mu^{ex}, \mu^{in} = 10^{-2} \mu^{ex}, \kappa^{in} = 10^3 \mu^{ex}, \epsilon^{in} = 10^2 \epsilon^{ex}$).

prior works have used pre-existing loads (or electrical fields) to engineer energy harvesting (Koh et al., 2011, 2009; Yang et al., 2017). Similar to those works, we consider the soft dielectric film filled with liquid inclusions subject to a uniform uniaxial external electric field through its thickness and in-plane equal biaxial load. Specifically we use a modified version of an analytical result by Koh et al. (2011) that relates the maximum energy of conversion to parameters such as dielectric constants, elastic properties, breakdown field among others. Essentially, Koh et al. (2011) derived the maximum energy of conversion for dielectric elastomers on the basis of four modes of failure: electrical breakdown, electromechanical instability, loss of tension, and material rupture. They focused on the full range of strain of operation, invoked a stress-strain relation that captures stiffening and also incorporated the fact that the electrical breakdown field is enhanced by strain. For the sake of ensuring durability, the authors investigated, among other things, the case of small strain operations (up to 5% stain). For materials verifying that $\epsilon^{ex} e_{EB}^2 < 6\mu^{ex} e_{max}$, they found the following analytical expression of the maximum energy of conversion Y_{max}

$$Y_{\text{max}} = \frac{1}{\rho} \left(2e_{\text{max}} \epsilon^{\text{ex}} e_{\text{EB}}^2 - \frac{(\epsilon^{\text{ex}} e_{\text{EB}}^2)^2}{6\mu^{\text{ex}}} \right)$$
(4.38)

such that e_{max} is the maximum strain of operation, e_{EB} is the magnitude of the electrical breakdown field and ρ is the mass density of 1000 kg m⁻³. In our context, of course, the permittivity must be replaced by suitable homogenized electrostrictive coefficients (and for that matter, elastic properties). For the geometry under consideration, the Maxwell stress component, according to (4.6), is $\sigma_{11} = \sigma_{22} = -\frac{1}{2} \epsilon^{\text{ex}} e_{\text{EB}}^2$. However, adding liquid inclusions leads to two important consequences. First, the dielectric elastomer film is not homogenous anymore, thus, we need to determine the corresponding effective Maxwell stress component. According to (4.10) and (4.11), we have $\sigma_{11}^{\text{eff}} = \sigma_{22}^{\text{eff}} = \mathbb{A}_{1133}^{\text{eff}} e_{\text{EB}}^2 = \mathbb{A}_{2233}^{\text{eff}} e_{\text{EB}}^2$. Consequently, we can replace ϵ^{ex} in their equations by $-2\mathbb{A}_{1133}^{\text{eff}}$ and μ^{ex} by μ^{eff} which leads to the following equation:

$$Y_{\text{max}} = \frac{1}{\rho} \left(-4e_{\text{max}} \mathbb{A}_{1133}^{\text{eff}} e_{\text{EB}}^2 - \frac{(2\mathbb{A}_{1133}^{\text{eff}} e_{\text{EB}}^2)^2}{6\mu^{\text{eff}}} \right)$$
(4.39)



Fig. 8. Maximum energy of conversion Y_{max} as a function of the volume fraction for only 5% operating strain. This plot shows the enhancement in the energy of conversion for a soft dielectric with liquid like inclusions as demonstrated in (4.39) such that $\kappa^{\text{ex}} = 10^3 \mu^{\text{ex}}$, $\mu^{\text{in}} = 10^{-2} \mu^{\text{ex}}$, $\kappa^{\text{in}} = 10^3 \mu^{\text{ex}}$, $\epsilon^{\text{in}} = 10^2 \epsilon^{\text{ex}}$, $e_{\text{EB}} = 30 \times 10^6$ V m⁻¹, $e_{\text{max}} = 0.05$, $\mu^{\text{ex}} = 0.5$ MPa, $\epsilon^{\text{ex}} = 3\epsilon_0$ and $\epsilon_0 = 8.854 \times 10^{-12}$ F m⁻¹.

Second, adding liquid inclusions into a soft dielectric polymer alters its electrical breakdown. As a matter of fact, in a recent study (Wang et al., 2012) it was shown that the breakdown electric field of a polymer filled with liquid drops scales as $Z\sqrt{\frac{\mu^{ex}}{\epsilon^{ex}}}$ where Z is a non-dimensional parameter that accounts for the geometry and interaction of the drops and approaches Z = 0.55 in case of a single drop. The aforementioned work not only concludes that soft dielectrics with higher moduli have a proportionally higher breakdown field, but also that increasing the volume fraction of liquid inclusions leads to a decrease of the breakdown electric field. Accordingly, in this part of the discussion, we restrict ourselves to a concentration not higher than 20% and believe that we can safely use a breakdown field $e_{EB} = 30 \times 10^6$ V m⁻¹ for the ensuing calculations.

Based on the stated arguments in the preceding paragraph, and in order to compare our results with the ones presented in Koh et al. (2011), we use the material parameters specified in the caption of Fig. 8.

In Fig. 8, when the volume fraction is 0, we recover the result for a homogeneous soft dielectric shown in Koh et al. (2011) where the maximum energy of conversion is estimated to be 2.2 mJ/g. Adding liquid inclusions and taking into account the capillary effect considerably enhances the performance of the composite as a generator. Even for only 5% of operating strain, we observe an improvement up to 74.9% for a 20% volume fraction and for a ratio $\gamma/(\mu^{ex}R_0) = 50$. This means that the smaller the radii of the liquid drops are (or larger the surface tension), the better the generator performance will be. For a soft dielectric elastomer with a shear modulus $\mu^{ex} = 0.5$ MPa, this value is achievable for nano-sized inclusions. We may further speculate that enhanced energy conversion is possible if we use a larger operating strain but a fully nonlinear framework (as opposed to linearized) must be used to assess those results.

4.5. Effective electrostrictive properties of composites without capillary effects and connection to past literature

As already discussed in prior sections, effective electrostrictive properties of composites (without consideration of capillarity) has been considered before. In this section, we simply present our results (stripped of capillarity, $\gamma = 0$) to make a connection with those works. Specifically, in Fig. 9 we compare the results with those of Spinelli et al. (2015). The effective properties without capillarity are

$$m_{K}^{\text{eff}} = \epsilon^{\text{ex}} + \Delta \epsilon^{2} \frac{\left(5 - 2v_{f} + \frac{15\epsilon^{\text{ex}}}{-\epsilon^{\text{ex}} + \epsilon^{\text{in}}}\right)}{15v_{f}\epsilon^{\text{ex}}} + \Delta \epsilon^{2} \Delta \mu \frac{1}{5v_{f}^{2}\epsilon^{\text{ex}}(\mu^{\text{ex}} - \mu^{\text{in}})},$$

$$m_{J}^{\text{eff}} = -\frac{\epsilon^{\text{ex}}}{2} - \frac{\Delta \epsilon}{2} + \Delta \epsilon^{2} \frac{3(1 - v_{f})(2\epsilon^{\text{ex}} + \epsilon^{\text{in}})(\kappa^{\text{ex}} - \kappa^{\text{in}})}{2v_{f}\epsilon^{\text{ex}}(\epsilon^{\text{ex}} - \epsilon^{\text{in}})(3v_{f}(\kappa^{\text{ex}} - \kappa^{\text{in}}) + 3\kappa^{\text{in}} + 4\mu^{\text{ex}})}.$$
(4.40)

Our results are only valid in the limit of incompressible inclusions and so we plot the results for a nearly incompressible dielectric elastomer filled with liquid-like high permittivity spherical particles such that $\kappa^{ex}/\mu^{ex} = 10^3$, $\mu^{in}/\mu^{ex} = 10^{-2}$, $\kappa^{in}/\mu^{ex} = 10^3$, $\epsilon^{in}/\epsilon^{ex} = 10^2$. Fig. 9 shows the normalized effective shear modulus μ^{eff}/μ^{ex} based on Eq. (3.30)₂, the permittivity $\epsilon^{eff}/\epsilon^{ex}$ using (4.18), the electrostrictive coefficients m_K^{eff}/ϵ^{ex} , $-2m_J^{eff}/\epsilon^{ex}$ based on Eq. (4.40) and the ratio of electrostriction \bar{E}_{33}/E_{33}^{ex} from (4.36) and (4.37), as a function of the volume fraction v_f . In these subfigures, the blue solid lines are the result of our theory. The turquoise dashed lines represent the Differential Coated Sphere (DCS) assemblage homogenization scheme (Lefevre and Lopez-Pamies, 2014). The red solid circles correspond to the finite element (FE) simulations results for isotropic suspensions of monodisperse spherical particles (Spinelli et al., 2015). We remark that our effective permittivity, shear modulus and electrostriction coefficients m_J^{eff} are consistent with the DCS and FE results. Thus, in absence of surface tension, the shear modulus decreases and both the normalized effective permittivity and the ratio



Fig. 9. Effective properties of a nearly incompressible dielectric elastomer filled with liquid-like high permittivity spherical particles in absence of capillarity. ($\kappa^{ex} = 10^3 \mu^{ex}, \mu^{in} = 10^{-2} \mu^{ex}, \kappa^{in} = 10^3 \mu^{ex}, \epsilon^{in} = 10^2 \epsilon^{ex}$). The blue solid lines are the result of our theory. The turquoise dashed lines represent the results for isotropic suspensions of polydispersed spherical particles for the differential coated sphere (DCS) assemblage (Lefevre and Lopez-Pamies, 2014). The red solid circles correspond to FE simulations results for isotropic suspensions of monodisperse spherical particles (Spinelli et al., 2015). The green dashed line stands for isotropic distribution of spherical particles with isotropic elastic dielectric properties (Spinelli et al., 2015). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

 $-2m_{J}^{\text{eff}}/\epsilon^{\text{ex}}$ increase as we add more liquid inclusions. However, when it comes the effective electrostrictive coefficient m_{K}^{eff} and the ratio $\bar{E}_{33}/E_{33}^{\text{ex}}$, we note a difference between the theoretical result of Spinelli et al. (2015) and ours, although our calculations do match their DCS and FE results.

5. Concluding remarks and prospects for future work

- (i) One of the key contribution of this work is to clarify the structure of the capillarity theoretical framework that is suitable for soft matter. While, the Gurtin and Murdoch (1975) framework is quite general, its linearized version is often used and interpreted in a manner that obfuscates physical insights underpinning the observation that the apparent "residual surface tension" can soften or enhance the stiffness of a soft solid depending on the capillary number.
- (ii) We find that the "surface tension" like term gives rise to an *apparent* surface elasticity tensor that is distinct from the surface elasticity tensor of the Gurtin–Murdoch theory that is frequently used in the context of hard solids.
- (iii) From the elaborated calculation in Section 3.1, one can see that the shape of liquid inclusions is qualitatively important. For non-spherical inclusions, we do expect "softening" effects of liquid inclusions, at least for some loading directions, even if the interior is incompressible. Spherical inclusions is peculiar because a spherical surface is a minimum surface for fixed interior volume. This aspect was not pursued in the present work but arguably represents an interesting extension.

(iv) Recognizing that in experiments, the liquid inclusions are frequently ionic in nature, we investigated the interplay between electrostatics, capillarity and high permittivity liquid inclusions for enhanced electrostriction and energy conversion in soft solids. We find that due to a competition between enhanced electrostriction and elastic stiffening, the actuator mode of the soft solid remains relatively unaffected by capillarity, its propensity for energy conversion is increased quite significantly. An interesting future extension could involved consideration of interfacial charges and/or *surface* electrostatics.

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Appendix A

A.1. Spherical coordinates and spherical harmonics

Let (r, θ, φ) be the usual spherical coordinates. First, we recall that axis-symmetric spherical harmonics are given by

$$Y_{l}(\theta) = \sqrt{\frac{2l+1}{4\pi}} P_{l}(\cos\theta), \qquad P_{l}(x) = \frac{1}{2^{l}l!} \frac{d^{l}}{dx^{l}} \left[(x^{2}-1)^{l} \right], \tag{A.1}$$

where $P_l: (-1, 1) \to \mathbb{R}$ is the Legendre polynomials satisfying

$$\frac{d}{dx}\left[(1-x^2)\frac{d}{dx}P_l(x)\right] + l(l+1)P_l(x) = 0.$$

Therefore, we have that $(\tilde{l} = l(l+1) \text{ and } ' = d/d\theta)$

 $r^2 \Delta Y_l = Y_l'' + \cot \theta Y_l' = -\tilde{l}Y_l,$

and the orthogonal properties (S is the unit spherical surface):

$$\int_{\mathbb{S}} Y_l Y_k da = \delta_{lk}, \quad \int_{\mathbb{S}} Y_l' Y_k' da = \tilde{l} \delta_{lk}, \quad \int_{\mathbb{S}} (\Delta_s Y_l) (\Delta_s Y_k) da = (\tilde{l})^2 \delta_{lk},$$

$$I_{lk} := \int_{\mathbb{S}} Y_l'' Y_k' \cot \theta da = -\int_{\mathbb{S}} Y_l' Y_k'' \cot \theta da + \int_{\mathbb{S}} Y_l' Y_k' da = -I_{kl} + \tilde{l} \delta_{kl}.$$
(A.2)

The axis-symmetric *vector* spherical harmonics are defined as $(\mathbf{e}_r = \mathbf{r}/r, \mathbf{e}_{\theta}, \mathbf{e}_{\varphi})$ are the unit basis vectors of spherical coordinate (r, θ, φ)

$$\mathbf{Y}_l = Y_l \mathbf{e}_r, \qquad \mathbf{\Psi}_l = r \nabla Y_l = Y_l' \mathbf{e}_\theta, \qquad \mathbf{\Phi}_l = \mathbf{r} \times \nabla Y_l.$$

It is convenient to explicitly write down l = 0, 2 modes:

$$Y_{0}(\theta) = \sqrt{\frac{1}{4\pi}}, \quad Y_{2}(\theta) = \frac{1}{4}\sqrt{\frac{5}{\pi}} \left(3\cos^{2}\theta - 1\right), \quad Y_{2}'(\theta) = -\frac{3}{4}\sqrt{\frac{5}{\pi}}\sin 2\theta.$$
(A.3)

Vector spherical harmonics being a complete basis, an axis-symmetric vector field admits the following expansion:

$$\mathbf{u}(\mathbf{x}) = u^{r}(r,\theta)\mathbf{e}_{r} + u^{\theta}(r,\theta)\mathbf{e}_{\theta} = \sum_{l=0}^{\infty} (X_{l}(r)\mathbf{Y}_{l} + \Theta_{l}(r)\mathbf{\Psi}_{l}),$$
(A.4)

where by (A.2),

$$X_{l}(r) = \frac{1}{r^{2}} \int_{\partial B_{r}} u^{r}(r,\theta) Y_{l}(\theta) da, \qquad \Theta_{l}(r) = \frac{1}{\tilde{l}r^{2}} \int_{\partial B_{r}} u^{\theta}(r,\theta) Y_{l}'(\theta) da.$$
(A.5)

Recall the transformation matrix between basis vectors of the spherical coordinates and the cartesian coordinate basis vectors:

$$\begin{bmatrix} \mathbf{e}_{x} \\ \mathbf{e}_{y} \\ \mathbf{e}_{z} \end{bmatrix} = \begin{bmatrix} \sin\theta\cos\phi & \cos\theta\cos\phi & -\sin\phi \\ \sin\theta\sin\phi & \cos\theta\sin\phi & \cos\phi \\ \cos\theta & -\sin\theta & 0 \end{bmatrix} \begin{bmatrix} \mathbf{e}_{r} \\ \mathbf{e}_{\theta} \\ \mathbf{e}_{\phi} \end{bmatrix}.$$
(A.6)

We now consider the axis-symmetric displacement $\mathbf{\bar{u}} = \mathbf{\bar{E}x} = (ax_1, ax_2, bx_3)$ on $\partial \Omega_0 = \{r = R_0\}$. By (A.5) and (A.6), we find that

$$\bar{\mathbf{u}} = \bar{X}_0 \mathbf{Y}_0 + \bar{X}_2 \mathbf{Y}_2 + \bar{\Theta}_2 \boldsymbol{\Psi}_2,$$

where

$$\bar{X}_0 = \frac{2}{3}R_0\pi^{1/2}(2a+b), \quad \bar{X}_2 = -\frac{4}{15}R_0(5\pi)^{1/2}(a-b) \quad \bar{\Theta}_2 = -\frac{2}{15}R_0(5\pi)^{1/2}(a-b). \tag{A.7}$$

Moreover, for an axis-symmetric vector field \mathbf{u}' on $\partial \Omega_0$ given by

$$\mathbf{u}'(\theta) = X_0' \mathbf{Y}_0(\theta) + X_2' \mathbf{Y}_2(\theta) + \Theta_2' \mathbf{\Psi}_2(\theta), \tag{A.8}$$

By (A.3) we find that $(\mathbf{n} = \mathbf{e}_r)$

$$\mathbf{u}' \otimes \mathbf{n} = \begin{bmatrix} \frac{1}{2\sqrt{\pi}} X_0' + \frac{1}{8} X_2' (3\cos(2\theta) + 1)\sqrt{\frac{5}{\pi}} & 0 & 0\\ \frac{1}{4} (-3)\sqrt{\frac{5}{\pi}} \Theta_2' \sin(2\theta) & 0 & 0\\ 0 & 0 & 0 \end{bmatrix}$$
(A.9)

with respect to the spherical coordinate system $\{\mathbf{e}_r, \mathbf{e}_{\theta}, \mathbf{e}_{\varphi}\}$. By (A.6) and direct integration we find that

$$\bar{\mathbf{E}}' := \frac{1}{4\pi R_0^2} \int_{\partial\Omega_0} \mathbf{u}' \otimes \mathbf{n} = \begin{bmatrix} \frac{5X_0' - \sqrt{5}(3\Theta_2' + X_2')}{30\sqrt{\pi}} & 0 & 0\\ 0 & \frac{5X_0' - \sqrt{5}(3\Theta_2' + X_2')}{30\sqrt{\pi}} & 0\\ 0 & 0 & \frac{5X_0' + 2\sqrt{5}(3\Theta_2' + X_2')}{30\sqrt{\pi}} \end{bmatrix},$$
(A.10)

where the matrix components are with respect to Cartesian coordinates (\mathbf{e}_x , \mathbf{e}_y , \mathbf{e}_z).

A.2. Determination of bulk elastic energies

The purpose of this subsection is to find the exterior and interior bulk elastic energies as determined by the boundary value problem:

$$\begin{cases} \operatorname{div}(\mathbb{C}_{\#}\nabla\mathbf{v}) = 0 & \text{in} \quad \{r < R_0\} \cup \{R_0 < r < R\}, \\ \mathbf{v} = \mathbf{v}^b & \text{on} \quad \{r = R_0\}, \\ \int_{\{r=R\}} \mathbf{v} \otimes \mathbf{n} = 0, \end{cases}$$
(A.11)

where $r < R_0$ (resp. $R_0 < r < R$) represents the inclusion (resp. matrix) region (see Fig. 2(a)), and \mathbf{v}^b is the prescribed interfacial displacement defined on $\partial \Omega_0$. By (A.4), a solution to (A.11) can in general be written as

$$\mathbf{v}(r,\theta) = \sum_{l=0}^{\infty} (X_l(r)\mathbf{Y}_l + \Theta_l(r)\mathbf{\Psi}_l).$$
(A.12)

Assuming isotropic bulk stiffness tensor $\mathbb{C}_{\#}$ and by substituting (A.12) into (A.11), we obtain, for each mode *l*, ODEs for mode amplitudes ($X_l(r)$, $\Theta_l(r)$).

• For
$$l = 0$$
 mode, we write
 $\mathbf{v}(r, \theta) = X_0(r) \mathbf{Y}_0(\theta).$

By (A.11), we end up with the following system of equations to find the displacement's amplitude

$$\begin{cases} \left(r^2 X_0'(r)\right)' - 2X_0(r) = 0 & \text{if } r \in (0, R_0) \cup (R_0, R), \\ X_0(R_0) = X_0^b, & \int_{\{r=R\}} X_0(r) \mathbf{e}_r \otimes \mathbf{e}_r = 0, \end{cases}$$
(A.14)

(A.13)

where $X_0^b = X_0(r = R_0)$. Immediately, we find the solution in the interior and exterior domains as $(v_f = R_0^3/R^3)$

$$X_{0}(r) = \begin{cases} \frac{X_{0}^{b}}{R_{0}}r & \text{if } r < R_{0}, \\ \frac{-X_{0}^{b}v_{f}}{R_{0}(1-v_{f})}r + \frac{X_{0}^{b}}{(1-v_{f})}\frac{R_{0}^{2}}{r^{2}} & \text{if } R_{0} < r < R. \end{cases}$$
(A.15)

Therefore, the interior strain energy can be expressed as a quadratic form of X_0^b :

$$\frac{1}{2} \int_{\{r< R_0\}} \nabla \mathbf{v} \cdot \mathbb{C}^{\operatorname{in}} \nabla \mathbf{v} = \frac{3}{2} \kappa^{\operatorname{in}} R_0 (X_0^b)^2.$$
(A.16)

Similarly, the exterior strain energy can be written as

$$\frac{1}{2} \int_{\{R_0 < r < R\}} \nabla \mathbf{v} \cdot \mathbb{C}^{e_X} \nabla \mathbf{v} = \frac{R_0 (X_0^b)^2 (4\mu^{e_X} + 3\kappa^{e_X} \nu_f)}{2(1 - \nu_f)}.$$
(A.17)

From (A.16) and (A.17), we can immediately identify the matrices \mathbf{D}_0^{in} and \mathbf{D}_0^{ex} in (3.22).

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• Similarly, for l = 2 mode we have

$$\mathbf{v}(r,\theta) = X_2(r)\mathbf{Y}_2(\theta) + \Theta_2(r)\mathbf{\Psi}_2(\theta). \tag{A.18}$$

Let X_2^b and Θ_2^b be the boundary values of $X_2(r)$ and $\Theta_2(r)$ at $r = R_0$. Inserting (A.18) into (A.11)₁, we end up with the following ordinary differential equations for $(X_2(r), \Theta_2(r))$ on $r \in (0, R_0) \cup (R_0, R)$:

$$\begin{cases} [r^2 X_2'(r)]' - \frac{2(\lambda+5\mu)}{(\lambda+2\mu)} X_2(r) - \frac{6(\lambda+\mu)}{(\lambda+2\mu)} r \Theta_2'(r) + \frac{6(\lambda+3\mu)}{(\lambda+2\mu)} \Theta_2(r) = 0, \\ [r^2 \Theta_2''(r)]' - \frac{6(\lambda+2\mu)}{\mu} \Theta_2(r) + \frac{\lambda+\mu}{\mu} r X_2'(r) + \frac{2(\lambda+2\mu)}{\mu} X_2(r) = 0, \end{cases}$$
(A.19)

where (μ, λ) are the Lamé constants. The general solutions to these ODEs can be written as follows:

$$X_2(r) = \frac{c_1}{r^4} + \frac{c_2}{r^2} + c_3 r + c_4 r^3, \\ \Theta_2(r) = -\frac{c_1}{3r^4} + \frac{c_2 \mu}{r^2 (3\lambda + 5\mu)} + \frac{c_3 r}{2} + \frac{c_4 r^3 (5\lambda + 7\mu)}{6\lambda},$$
(A.20)

where, in order to keep it general, we are not specifying the superscript e^x or i^n to the Lamé constants, and c_i (i = 1, ..., 4) are constants to be determined by the last of (A.11) and the boundary conditions:

$$X_2(R_0) = X_2^b, \qquad \Theta_2(R_0) = \Theta_2^b.$$
 (A.21)

More specifically, for the interior region $\{r < R_0\}$ we infer that $c_1 = c_2 = 0$ and, by (A.21), determine c_3 , c_4 in terms of X_2^b, Θ_2^b :

$$c_{3}^{\text{in}} = \frac{(\Theta_{2}^{b}(12\mu^{\text{in}} - 18\kappa^{\text{in}}) + X_{2}^{b}(15\kappa^{\text{in}} + 11\mu^{\text{in}}))}{6\kappa^{\text{in}}R_{0} + 17\mu^{\text{in}}R_{0}}, c_{4}^{\text{in}} = -\frac{3r^{3}(3\kappa^{\text{in}} - 2\mu^{\text{in}})(X_{2}^{b} - 2\Theta_{2}^{b})}{R_{0}^{3}(6\kappa^{\text{in}} + 17\mu^{\text{in}})}.$$
(A.22)

Similarly, for the exterior region $\{r > R_0\}$ we infer that $c_4 = 0$ by the far field strain condition $(\nabla \mathbf{v} \to 0 \text{ at } r \to \infty)$ and, by (A.21) and the last of (A.11), determine c_1, c_2, c_3 in terms of X_2^b, Θ_2^b :

$$\mathcal{L}_{1}^{ex} = \frac{R_{0}^{4}(15\Theta_{2}^{b}(\kappa^{ex} + \mu^{ex}) + 3\nu_{f}(\kappa^{ex} + 2\mu^{ex})(X_{2}^{b} - 2\Theta_{2}^{b}) - 5\mu^{ex}X_{2}^{b})}{5(\nu_{f} - 1)(\kappa^{ex} + 2\mu^{ex})}, \\
\mathcal{L}_{3}^{ex} = \frac{2r\nu_{f}(3\Theta_{2}^{b} + X_{2}^{b})}{5R_{0}(\nu_{f} - 1)}, \quad (A.23)$$

Therefore, by (A.20), (A.22), (A.23) we find that the interior strain energy for this mode can be written as

$$\frac{1}{2} \int_{\{r < R_0\}} \nabla \mathbf{v} \cdot \mathbb{C}^{\text{in}} \nabla \mathbf{v} = \frac{6\Theta_2^2 \mu^{\text{in}} R_0 (15\kappa^{\text{in}} + 11\mu^{\text{in}})}{6\kappa^{\text{in}} + 17\mu^{\text{in}}} + \frac{\mu^{\text{in}} R_0 X_2^2 (39\kappa^{\text{in}} + 16\mu^{\text{in}})}{12\kappa^{\text{in}} + 34\mu^{\text{in}}} + \frac{18\Theta_2 \mu^{\text{in}} R_0 X_2 (2\mu^{\text{in}} - 3\kappa^{\text{in}})}{6\kappa^{\text{in}} + 17\mu^{\text{in}}},$$
(A.24)

and the exterior strain energy can be written as

$$\frac{1}{2} \int_{r>R_0} \nabla \mathbf{v} \cdot (\mathbb{C}^{ex} \nabla \mathbf{v}) = -\frac{1}{2} \int_{\{r=R_0\}} \mathbb{V} \cdot (\mathbb{C}^{ex} \nabla \mathbf{v}) \mathbf{n} \\
= \frac{3\Theta_2^2 \mu^{ex} R_0(\kappa^{ex}(2\nu_f - 5) + 4\mu^{ex}(\nu_f - 2))}{(\nu_f - 1)(\kappa^{ex} + 2\mu^{ex})} + \frac{\mu^{ex} R_0 X_2^2 (\kappa^{ex}(6\nu_f - 9) + 4\mu^{ex}(3\nu_f - 4))}{3(\nu_f - 1)(\kappa^{ex} + 2\mu^{ex})} \\
+ \frac{2\Theta_2 \mu^{ex} R_0 X_2 (\kappa^{ex}(3 - 6\nu_f) + 4\mu^{ex}(2 - 3\nu_f))}{(\nu_f - 1)(\kappa^{ex} + 2\mu^{ex})}.$$
(A.25)

From (A.24) and (A.25), we can identify the matrices \mathbf{D}_2^{in} and \mathbf{D}_2^{ex} in (3.22).

A.3. Electrostrictive stress in the spherical coordinate system

Here, we are going to use the solution of the potential found in (4.17) and plug it directly into the coupled problem (4.23). From (4.5) and (4.6), we will first to calculate the interior and exterior electric field on $\partial \Omega_0$:

$$\nabla \Phi^{\text{in}} = \begin{bmatrix} -c_0^{\text{in}} e_0 \cos\theta \\ c_0^{\text{in}} e_0 \sin\theta \\ 0 \end{bmatrix}, \qquad \nabla \Phi^{\text{ex}} = \begin{bmatrix} (-c_1^{\text{ex}} - 2c_2^{\text{ex}})e_0 \cos\theta \\ (c_1^{\text{ex}} + c_2^{\text{ex}})e_0 \sin\theta \\ 0 \end{bmatrix}.$$
(A.26)

Therefore, by (4.5) we find the jump of the electrostrictive stress across the interface $\partial \Omega_0$:

$$\llbracket \boldsymbol{\sigma}_{\text{elect}} \rrbracket = \left(\boldsymbol{\sigma}_{\text{elect}}^{\text{ex}} - \boldsymbol{\sigma}_{\text{elect}}^{\text{in}} \right) \Big|_{r=R_0}$$
(A.27)

with relevant nonzero components given by

$$(\llbracket \boldsymbol{\sigma}_{\text{elect}} \rrbracket)_{rr} = \frac{9e_0^2 \epsilon^{\text{ex}} (\epsilon^{\text{ex}} - \epsilon^{\text{in}}) (-\epsilon^{\text{ex}} - \epsilon^{\text{in}} + (\epsilon^{\text{ex}} - \epsilon^{\text{in}}) \cos 2\theta)}{4((v_f + 2)\epsilon^{\text{ex}} - v_f \epsilon^{\text{in}} + \epsilon^{\text{in}})^2},$$

$$(\llbracket \boldsymbol{\sigma}_{\text{elect}} \rrbracket)_{\theta\theta} = \frac{9e_0^2 \epsilon^{\text{ex}} (\epsilon^{\text{ex}} \sin^2 \theta + \epsilon^{\text{in}} \cos 2\theta) - (\epsilon^{\text{in}})^2 \cos^2 \theta)}{2((v_f + 2)\epsilon^{\text{ex}} - v_f \epsilon^{\text{in}} + \epsilon^{\text{in}})^2}$$

By (A.27) and (A.9), we obtain (4.22).

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