Interfacial tension and a three-phase generalized self-consistent theory of non-dilute soft composite solids

Francesco Mancarella,1 Robert W. Style,2 and John S. Wettlaufer3,2,1
1Nordic Institute for Theoretical Physics (NORDITA), SE-106 91 Stockholm, Sweden
2Mathematical Institute, University of Oxford, Oxford OX1 3LB, UK
3Yale University, New Haven, Connecticut 06520, USA
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In the dilute limit Eshelby’s inclusion theory captures the behavior of a wide range of systems and properties. However, because Eshelby’s approach neglects interfacial stress, it breaks down in soft materials as the inclusion size approaches the elastocapillarity length \( L \). Here, we use a three-phase generalized self-consistent method to calculate the elastic moduli of composites comprised of an isotropic, linear-elastic compliant solid hosting a spatially random monodisperse distribution of spherical liquid droplets. As opposed to similar approaches, we explicitly capture the liquid-solid interfacial stress when it is treated as an isotropic, strain-independent surface tension. Within this framework, the composite stiffness depends solely on the ratio of the elastocapillarity length \( L \) to the inclusion radius \( R \). Independent of inclusion volume fraction, we find that the composite is stiffened by the inclusions whenever \( R < 3L/2 \). Over the same range of parameters, we compare our results with alternative approaches (dilute and Mori-Tanaka theories that include surface tension). Our framework can be easily extended to calculate the composite properties of more general soft materials where surface tension plays a role.

I. INTRODUCTION

Composite materials are of interest because they rarely have the bulk properties of their constituents alone. Thus, understanding their properties provides a challenge and a test bed for both controlling material behavior and understanding how and why natural materials have evolved [e.g., 1, 2, and refs. therein]. Because in both engineering and natural settings there is always a compromise between the ability to tailor every detail and achieving an optimal effective behavior, such as stiffness, theoretical approaches that span the widest range of key control parameters are desirable.

Among the most successful idealized geometric models for two-phase matrix-inclusion composites is Hashin’s composite-spheres model [3, 4], where the actual composite is replaced by a set of “composite spheres” with a suitable size-distribution, and arranged in a volume-filling configuration. Each composite sphere consists of a homogeneous sphere representing the inclusion phase, surrounded by a concentric spherical shell of matrix material. The ratio between internal and external radii of each shell is determined in terms of the volume fraction occupied by the inclusion phase within the actual composite. While the effective bulk modulus is determined analytically, there is no exact solution for the effective shear modulus, although considerable information is available on its variational bounds.

In addition to the bounds mentioned above, a broad class of self-consistent (SC) methods have been developed that yield analytical predictions for both bulk and shear moduli. For example, Kroner [5] introduced a self-consistent approximation wherein the inclusion itself is directly embedded in an unknown homogeneous effective medium. Budiansky [6] and Hill [7, 8] use this approach in their model for elastic moduli of composites, although they noted physical inconsistencies at high inclusion volume fractions \( \phi \).

A three-phase generalized self-consistent (GSC) model was introduced by Kern [9, 10] and van der Poel [11]. Christensen and Lo [12] took this approach by replacing the set of all actual inclusions by a single ideal inclusion, in the “composite spheres” framework described above. Whereas the SC method is generally simpler than GSC models, the proper boundary conditions of the latter remove the unphysical behavior of the former as \( \phi \) becomes large. Both approaches typically assume continuity of strain (or no slip) across interfaces [e.g., 13], although the bulk modulus has been shown to be unaffected by finite slip [14].

The surface stress at solid/liquid interfaces can have a substantial range of size-dependent effects in soft materials. For example, recent work has shown that surface stress significantly influences pearling and creasing instabilities [15–18], wetting [19–24], adhesion [25,28], and the relaxation of soft solids towards their equilibrium shapes [e.g., 29]. Hence, here we aim to reformulate the micromechanics of soft composites in the non-dilute limit to include the effects of surface stress. In so doing, we systematically examine how the interplay between the inclusion volume fraction \( \phi \) and the inclusion size \( R \) influences the mechanical properties. We note that, for both dilute and non-dilute cases, the inclusion/matrix surface stress has been treated in previous work, the most notable of which however assumes linear (in-strain) surface stress [30–34], and/or use incorrect boundary conditions, as described previously [35].

Style et al., [35, 36] studied a dilute monodisperse random spatial distribution of liquid droplets of radius \( R \) embedded in a homogeneous isotropic elastic solid ma-
Homogeneous composite

Elastic matrix

Liquid

μ, ν2

μ, ν3

R

R/ϕ1/3

Surface tension γ

II. THE MODEL

Consider a composite system of many identical incompressible droplets embedded in an isotropic homogeneous elastic solid with shear modulus μ2 and Poisson ratio ν2 as shown in Fig. 1. We ask how surface tension at the droplet/matrix interface affects Christensen and Lo’s [12] solution for the effective modulus of the composite for non-dilute droplet volume fractions. Our approach is based on the three-phase self-consistent model of Kerner [9].

As noted above, the GSC approach treats the multi-droplet system as a single composite sphere embedded in an infinite medium of unknown effective elastic moduli μ3, ν3. The composite sphere consists of a liquid droplet of radius R, surrounded by a concentric spherical shell of matrix material of radius R/ϕ1/3, thereby preserving the liquid volume fraction ϕ of the original multi-droplet system. The overall approach follows that of previous work [e.g. 35, 38], up to a point. To make this paper reasonably self-contained we summarize the key intermediate results, and provide more detail where we believe clarity is required.

Placing the origin of an (r, θ, φ) spherical coordinate system at the center of the composite sphere, we choose the following far-field (r → ∞) displacements:

\[ u^0_r = 2ε^0_A r P_2(\cos θ), \quad u^0_θ = ε^0_A r \frac{dP_2(\cos θ)}{dθ}, \quad u^0_φ = 0, \]

where \( P_2 \) is the Legendre polynomial of order 2; the corresponding, purely deviatoric far-field strains are:

\[ ε^0_{xx} = ε^0_{yy} = -ε^0_A, \quad ε^0_{zz} = 2ε^0_A. \]

For the strained system, the symmetry about the z-axis allows the use of the following ansatz [e.g., 33, 35, 39] for the displacements \( u_r^{(i)} \) and \( u_θ^{(i)} \) in the radial and polar directions,

\[ u_r^{(i)}(ρ, θ) = \left( F_i + \frac{G_i}{ρ^3} \right) r + P_2(\cos θ) \times \left[ 12ν_i A_i ρ^2 + 2B_i + 2\left( \frac{5 - 4ν_i}{ρ^3} \right) C_i - 3\frac{D_i}{ρ^3} \right] r, \]

\[ u_θ^{(i)}(ρ, θ) = \frac{dP_2(\cos θ)}{dθ} × \left[ (7 - 4ν_i) A_i ρ^2 + 2B_i + 2\left( \frac{1 - 2ν_i}{ρ^3} \right) C_i + \frac{D_i}{ρ^3} \right] r, \]

where \( ρ \equiv r/R \), the index "i" refers to either the matrix \( i = 2 \) or the composite effective medium \( i = 3 \) phase, and \( A_i \) through \( G_i \) will be determined from the boundary conditions. The corresponding stress components in regions \( i = 2, 3 \) are

\[ σ_r^{(i)}(ρ, θ) = 2μ_i \left( -2\frac{G_i}{ρ^3} + \frac{F_i(1 + ν_i)}{1 - 2ν_i} + \frac{6ν_i A_i ρ^2 + 2B_i - 4(5 - ν_i)C_i + 2D_i}{ρ^3} P_2(\cos θ) \right), \]

and

\[ σ_{θθ}^{(i)}(ρ, θ) = 2μ_i \frac{dP_2(\cos θ)}{dθ} × \left[ (7 + 2ν_i) A_i ρ^2 + 2B_i + \frac{2(1 + ν_i)}{ρ^3} C_i + \frac{4D_i}{ρ^3} \right]. \]

The relation between the pressure \( p \) and the components of the hydrostatic stress tensor in the liquid region (i=1) is:

\[ σ^{(1)}_r = σ^{(1)}_θ = -p, \quad σ^{(1)}_θ = 0. \]

Now, combining the stress-displacement relationship with Eq. (1) gives the far-field stresses:

\[ σ_r^{(0)} = 4ε^0_A μ_3 P_2(\cos θ), \quad σ_{θθ}^{(0)} = 2ε^0_3 μ_3 \frac{dP_2(\cos θ)}{dθ}. \]

Three constants are determined by the far-field stress/strain, viz., \( A_3 = 0, B_3 = ε^0_3, \) and \( F_3 = 0. \)
There are ten equations for the remaining ten unknowns; \( p, \mathcal{A}_2, \mathcal{B}_2, C_2, D_2, F_2, G_2, C_3, D_3, G_3 \). Six equations arise from continuity of strain and stress at the composite sphere surface \( (\rho = a \equiv 1/\phi^{1/3}) \), one from the incompressibility of the droplet and three from the stress boundary conditions associated with the generalized Young-Laplace condition. Taking these in turn, we have

\[
\begin{align*}
&u_r^2(a, \theta) = u_0^3(a, \theta), \\
&\sigma_r(a, \theta) = \sigma_0^3(a, \theta), \\
&\sigma_r(a, \theta) = \sigma_0^3(a, \theta),
\end{align*}
\]  

where the continuity of strain for \( u_r(0) \) provides two (one) equations and that for stress \( \sigma_r(0) \) provides two (one) equations; droplet incompressibility that the last two sumsmands exactly cancel (this was not pointed out in previous work [35]).

The condition \( W = 0 \) provides a constraint in the form of a quadratic equation, which determines the relative effective shear modulus \( \mu_{rel} / \mu_2 \). Indeed, up to second order in \( u \), we can replace the normal vector \( n \) in (13) by the basis unit vector \( \hat{r} \) of the spherical coordinate system and find that

\[
W = \frac{1}{2} \int_{S_{int}} \left[ \sigma_{rr} u_r + \sigma_{\theta r} u_\theta - \sigma_{r \theta} u_r - \sigma_{rr} u_\theta \right] dS = -\frac{48\pi \mu_3 R^3 \varepsilon_0^0(v_3 - 1)}{a^2} C_3.
\]  

Therefore, \( C_3 = 0 \), and plugging this into the solution of the system of equations (9)-(12) yields a quadratic condition for the relative effective shear modulus \( \mu_{rel} \equiv \mu_3 / \mu_2 \) as a function of \( \phi, v_2, \) and \( f/(R_{rel}) \) as follows

\[
2R_{rel}(a + a_1 \mu_{rel} + a_2 \mu_{rel}^2) + \gamma(b_0 + b_1 \mu_{rel} + b_2 \mu_{rel}^2) = 0,
\]  

where the coefficients are in Appendix A.

For the remainder of the paper we will focus on the special case of an incompressible matrix, for which \( E_{rel} \equiv \left( \frac{E}{E_2} \right) = \left( \frac{\mu_2}{\mu_2} \right) \equiv \mu_{rel} \) and \( v_2 = 1/2 \). Considering the elastocapillarity length \( L \equiv \gamma / E_2 \) based on the matrix phase of the composite sphere, we define the dimensionless parameter \( \gamma' \equiv L/R = \gamma/(E_2R) \). Fig. 2 shows the behavior of \( E_{rel} \) as a function of \( \phi \), and in Fig. 3 \( E_{rel} \) is plotted against \( R/(3V/4\pi)^{1/3} \), where \( V \) is the outer sphere volume in the GSC framework (i.e., \( E_{rel} \) is plotted against \( \phi^{1/3} \)). Clearly, Fig. 2 shows monotonic response over a large range of \( \phi \), exhibiting softening (stiffening) \( \gamma' < 2/3 \) \( (\gamma' > 2/3) \) behavior that spans the experimental range seen by Style et al. [36]. Moreover, the dilute theory [35] is quantitatively captured in the limit \( \phi \to 0 \) of the present theory. Furthermore, we find exact “mechanical cloaking”, in which \( E_{rel} \) is constant at \( \gamma' = 2/3 \) for all liquid volume fractions. Exactly the same cloaking condition is found in the dilute theory [35], and from a complimentary Mori-Tanaka approach [37] as described in Fig. 4 below.

In the stiffening regime, as droplets become small and \( \gamma' \) becomes large, the quadratic condition (15) for \( \mu_{rel} \) simplifies to \( b_0 + b_1 \mu_{rel} + b_2 \mu_{rel}^2 = 0 \). At a given \( \phi \) the solution of this equation, \( \mu_{rel} = \mu_{rel}(R \equiv \left( \phi, v_2 \right)) \), gives the upper limit of rigidity among all \( \gamma' \)-curves, showing a stiffening behavior proportional to \( \frac{1}{\phi^{1/3}} \) in the limit \( \phi \to 0 \) \( (\gamma' \to \infty \text{-line in Fig. 2}) \).

Now we examine the deformation of the inclusion phase by making use of Eqs. (B1) and (B2) in Appendix B to evaluate the effective droplet strain \( \varepsilon_d \equiv (l - 2R)/R = 2u_0(1, 0)/R \), where \( l \) denotes the major axis of the droplet[50]. In terms of \( \alpha = \phi^{-1/3}, \gamma' \), and the solution of Eq.(15), \( \mu_{rel} = \mu_3 / \mu_2 \), the radial and polar displacements of the droplet interface are

\[
\frac{u_r(1, \theta)}{R} = 100 \mu_{rel} \varepsilon_0 \frac{f_1}{f_2 + \gamma' f_3} \mathcal{P}_2(\cos \theta)
\]  

where \( \mathcal{P}_2 \) is the even Legendre polynomial of degree 2.
where the coefficients $f_1 - f_5$ are in Appendix B. When
$R \ll L$ and $\gamma' \ll 1$, and the radial displacement becomes
extremely small, then the inclusions remain spherical. In
the opposite limit, $R \gg L$ and $\gamma' \ll 1$, the inclusions
are compared quantitatively with the dilute theory and
in agreement with the theory for the case of pure bulk elasticity. The corresponding
effective droplet strain is $\varepsilon_d = 200\mu_{rel}\alpha^2\varepsilon_d^0 / (f_1 + \gamma' f_3)$. In
the dilute limit $\phi \to 0$ of the incompressible case, the
droplet’s effective strain and shape reduce to $\varepsilon_{d,\phi \to 0} = 40\varepsilon_d^0 / (6 + 15\gamma')$,
\[
\frac{u_0(1, \theta)}{R} = \frac{5}{3} \epsilon_0 \frac{1 + 3 \cos(2 \theta)}{2 + 5 \gamma'},
\]
and
\[
\frac{u_0(1, \theta)}{R} \bigg|_{\phi \to 0} = -\frac{5}{2} \epsilon_0 \frac{(2 + 3 \gamma') \sin(2 \theta)}{2 + 5 \gamma'},
\]
thereby recovering the results of Style et al. [35]. Interestingly, we find that in this limit, at the exact cloaking
point ($\gamma' = 2/3$), the droplet will stretch less than the
host material viz., $(1 - 2R)/(2R) = (5/8)\epsilon_0^0$. However,
the droplet will stretch the same as the host material at
$\gamma' = 4/15 < 2/3$, within the softening regime. The predictions of Eshelby’s theory [41] and effective droplet
strain $\varepsilon_{eff}^0$ are recovered for $\phi, \gamma' \ll 1$, whereas an
unperturbed spherical shape is found when $\gamma' \gg 1$, and
arbitrary values of $\phi$.

Finally, in Fig. 4, we compare $E_{rel}$ for this theory
(red) with a modified version of the Mori-Tanaka theory
(green [37]) and the dilute theory (blue [35]). We see
that this theory predicts a more pronounced softening in
the softening regime ($\gamma' < 2/3$), and a more pronounced
stiffening in the stiffening regime ($\gamma' > 2/3$) than does
the modified Mori-Tanaka theory. Interestingly, in the
stiffening regime, the three-phase and dilute theories are
perhaps experimentally indistinguishable, well beyond the
concentration range where the latter breaks down ($\phi \approx 0.2$). This indicates that, depending on the range
of $\gamma'$ of relevance, the dilute theory provides a simple
framework for comparison with experiment given that
it is the appropriate asymptotic limit of the non-dilute
theory. All three theories predict exactly the same me-
chanical cloaking condition, $\gamma' = 2/3$, of the inclusions,
independent of $\phi$. We note here that the results in the
surface-tension free limit $\gamma \to 0$ are compared with the
classical result [12] in Appendix A.

III. CONCLUSIONS

Based on a three-phase generalized self-consistent ap-
proach, we have estimated elastic moduli of com-
posites including liquid droplets, by taking into account the
(linear-elastic) solid/droplet interfacial surface tension.
In the limit $\phi \to 0$, we recover the dilute-theory expres-
sions of Style et al., [35, 36]. The Young’s modulus of the
composite depends on $\gamma'$, which is the ratio of the elasto-
capillary length $L$, to the inclusion radius $R$. The results
are compared quantitatively with the dilute theory and
a version of Mori-Tanaka theory, both of which include

FIG. 2: $E_{rel}$ against $\phi$ over a wide range of $\gamma'$ (as labeled in the figure) from the softening to the stiffening regime for the incompressible case $\nu_2 = 1/2$.

FIG. 3: $E_{rel}$ against $R/[(3V/4\pi)^{1/3}]$ for a wide range of the parameter $L/[(3V/4\pi)^{1/3}]$ (as labeled in the figure) for the incompressible case $\nu_2 = 1/2$.
Finally, we note that there is an interesting similarity between the mechanical response of the multi-phase soft materials studied here and what one finds in poroelasticity, which is a framework used to study the effective medium response of fluid filled host structures, applied to problems ranging from biology to geophysics [e.g., 42–47]. For example, in many biological settings, the composite medium has soft elastic or liquid inclusions, and the deformation of the host material is controlled by the value of $\phi$, which is typically determined as part of the solution to the problem. Whereas in poroelasticity a major challenge involves the modeling of the flow permeability, which is specified as a function of $\phi$, our approach derives the mechanical response as a function of $\phi$. We suggest that by treating the mechanical properties of poroelastic media within the framework studied here, one can constrain the $\phi$ dependence of transport properties such as the flow permeability.

**IV. ACKNOWLEDGMENTS**

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Appendix A: Coefficients in Equation (15)

The coefficients in equation (15) are

\[
\begin{align*}
\begin{cases}
  a_0 &= -49 - 252\alpha^3 + 25\nu_2^3 - 25\alpha^7 - 7 + 25\nu_2^3 - \alpha^{10}(49 + 25\nu_2^3) \\
  a_1 &= -7 + 504\alpha^3 + 30\nu_2 + 150\alpha^7 - 3 + 25\nu_2 + 50\alpha^7 - 7 + 25\nu_2^3 - 3\alpha^{10}(49 - 140\nu_2 + 75\nu_2^2) \\
  a_2 &= 56 - 252\alpha^3 + 30\nu_2 - 50\nu_2^3 - 25\alpha^7 + 25\alpha^7 - 7 - 12\nu_2 + 8\nu_2^2 + 4\alpha^{10}(49 - 105\nu_2 + 50\nu_2^2)
\end{cases}
\end{align*}
\]

and

\[
\begin{align*}
\begin{cases}
  b_0 &= 252\alpha^3(1 + 2\nu_2) + 25\alpha^7(7 + 2\nu_2) - 50\alpha^3(7 + 6\nu_2 + 4\nu_2^3) + 4(49 - 63\nu_2 + 20\nu_2^2) + \alpha^{10}(-119 + 48\nu_2 + 95\nu_2^2) \\
  b_1 &= -150\alpha^7(-3 + 2\nu_2)\nu_2 - 504\alpha^3(-1 + 2\nu_2) + 100\alpha^7(-7 + 6\nu_2 + 4\nu_2^3) + 4(7 - 39\nu_2 + 20\nu_2^2) - 3\alpha^{10}(119 - 388\nu_2 + 285\nu_2^2) \\
  b_2 &= 2(126\nu_3(-1 + 2\nu_2) - 25\alpha^7(-6 + 6\nu_2 + 4\nu_2^3) + 25\alpha^7(7 - 12\nu_2 + 8\nu_2^2) - 4(28 - 51\nu_2 + 20\nu_2^2) + \alpha^{10}(238 - 606\nu_2 + 380\nu_2^2))
\end{cases}
\end{align*}
\]

We note here that Eq. (3.14) of Christensen and Lo [12] is incorrect in the regime of large droplets \((R \gg L)\). In this regime, the condition (15) reduces to \(a_0 + a_1\mu_{rel} + a_2\mu_{rel}^2 = 0\), whose solution \(\mu_{rel} = \mu_{rel,R\gg L}(\theta, \nu_2)\) is invariant under \(\mu_2\)-scalings, as expected from the symmetries of the equations of the elastostatics.

Appendix B: Shape of the droplets under uniaxial stress

By using our model, we determine the shape of the generic droplet embedded in an incompressible matrix \((\nu_2 = 1/2)\) undergoing uniaxial stress. Note that, with \(c_{11}^0 = c_{22}^0/2\), the purely deviatoric far-field strain conditions of Eqs. (2) are equivalent to the strain system of Style et al., [35, see 3 lines below Eq.(2)]. Substituting the expressions for \(\mathcal{A}_2 \cdot \mathcal{G}_2\) into Eqs. (3) and (4), we obtain the following surface displacements,

\[
\frac{u_r(1, \theta)}{R} = [6\mathcal{A}_2 + 2\mathcal{B}_2 + 6\mathcal{C}_2 - 3\mathcal{D}_2] \mathcal{P}_2(\cos \theta) , \quad \text{(B1)}
\]

Finally, we note that the coefficients \(f_i\) in Eqs. (16) and (17) are:

\[
\begin{align*}
\begin{cases}
  f_1 &= 40(\mu_{rel} - 1) - 21(\mu_{rel} - 1)\alpha^2 + (19 + 16\mu_{rel})\alpha^7 \\
  f_2 &= 6(38(\mu_{rel} - 1)^2 + 75(\mu_{rel} - 1)(2 + 3\mu_{rel})\alpha^7 + 112(2 + \mu_{rel} - 3\mu_{rel}^2)\mu_5^7 + 50(\mu_{rel} - 1)(3 + 4\mu_{rel})\alpha^7 + (2 + 3\mu_{rel})(19 + 16\mu_{rel})\alpha^{10}) \\
  f_3 &= 15(-48(\mu_{rel} - 1)^2 + 40(\mu_{rel} - 1)(2 + 3\mu_{rel})\alpha^7 + 30(3 + \mu_{rel} - 4\mu_{rel}^2)\alpha^7 + (2 + 3\mu_{rel})(19 + 16\mu_{rel})\alpha^{10}) \\
  f_4 &= 10(\mu_{rel} - 1) + 28(\mu_{rel} - 1)\alpha^2 + 2(19 + 16\mu_{rel})\alpha^7 \\
  f_5 &= 3(40(\mu_{rel} - 1) - 56(\mu_{rel} - 1)\alpha^2 + (19 + 16\mu_{rel})\alpha^7)
\end{cases}
\end{align*}
\]

\[
\frac{u_\theta(1, \theta)}{R} = \left[5\mathcal{A}_2 + 3\mathcal{B}_2 + \mathcal{D}_2\right] \frac{d\mathcal{P}_2(\cos \theta)}{d\theta} . \quad \text{(B2)}
\]

\[
\begin{align*}
\begin{cases}
  f_1 &= -20(\mu_{rel} - 1) - 7(\mu_{rel} - 1)\alpha^2 + (9 + 16\mu_{rel})\alpha^7 \\
  f_2 &= 4(38(\mu_{rel} - 1)^2 + 75(\mu_{rel} - 1)(2 + 3\mu_{rel})\alpha^7 + 112(2 + \mu_{rel} - 3\mu_{rel}^2)\mu_5^7 + 50(\mu_{rel} - 1)(3 + 4\mu_{rel})\alpha^7 + (2 + 3\mu_{rel})(19 + 16\mu_{rel})\alpha^{10}) \\
  f_3 &= 15(-48(\mu_{rel} - 1)^2 + 40(\mu_{rel} - 1)(2 + 3\mu_{rel})\alpha^7 + 30(3 + \mu_{rel} - 4\mu_{rel}^2)\alpha^7 + (2 + 3\mu_{rel})(19 + 16\mu_{rel})\alpha^{10}) \\
  f_4 &= 10(\mu_{rel} - 1) + 28(\mu_{rel} - 1)\alpha^2 + 2(19 + 16\mu_{rel})\alpha^7 \\
  f_5 &= 3(40(\mu_{rel} - 1) - 56(\mu_{rel} - 1)\alpha^2 + (19 + 16\mu_{rel})\alpha^7)
\end{cases}
\end{align*}
\]

References:

[1] L. A. Mihai, K. Alayyash and A. Goriely, *Proceedings of the Royal Society of London A*, 2015, 471, 20150107.
[2] C. Coulais, J. T. B. Overvelde, L. A. Lubbers, K. Bertoldi and M. van Hecke, *Phys. Rev. Lett.*, 2015, 115, 044301.
[3] Z. Hashin and W. Rosen, *J. Appl. Mech.*, 1964, 31, 223-232.
[4] Z. Hashin, *J. Appl. Mech., Trans. ASME*, 1962, 29, 143-150.
[5] E. Kröner, *Zeitschrift für Physik*, 1958, 151, 504-518.
[6] B. Budiansky, *J. Mech. Phys. Solids*, 1965, 13, 223-227.
[7] R. Hill, *J. Mech. Phys. Solids*, 1965, 13, 213-222.
[8] R. Hill, *J. Mech. Phys. Solids*, 1965, 13, 189-198.
[9] E. H. Kerner, *Proc. Phys. Soc. B*, 1956, 69, 808-813.
[10] see also e.g. ref. 12 or refs. 48, 49, although unjustified assumptions are invoked in these two latter papers. Note further that the relative effective shear modulus formula \(\mu/\mu_n\) presented in ref. 12 (Eqs. 3.14 through 3.18) should be revised. This is evident from its symmetry breaking prop-
tery under arbitrary rescaling of the matrix shear modulus \( \mu_m \), whereas \( \mu / \mu_m \) is expected to be invariant in the case of (interface stress-free) liquid inclusions. Such a symmetry breaking is clear as \( \eta_2 \) in their Eq. (3.18), and hence also the solution \( \mu / \mu_m \) of the quadratic Eq. (3.14), is \( \mu_m \) - dependent even in the liquid inclusion limit \( \mu_l \to 0 \).

[11] C. van der Poel, *Rheol. Acta*, 1958, 1, 198-205.

[12] R. M. Christensen and K. H. Lo, *J. Mech. Phys. Solids*, 1979, 27, 315-330.

[13] R. A. Shick and H. Ishida, in *Characterization of Composite Materials*, ed. Hatsuo Ishida, Momentum Press LLC, New York, reprint edition, 2010, Chp.8, pp.148-183.

[14] K. Takahashi, M. Ikeda, K. Harakawa, K. Tanaka and T. Sakai, *J. Polym. Sci.: Polym. Phys.*, 1978, 16, 415-425.

[15] S. Mora, T. Phou, J.-M. Fromental, L. M. Pismen and Y. Pomeau, *Phys. Rev. Lett.*, 2010, 105, 214301.

[16] S. Mora, M. Abkarian, H. Tabuteau and Y. Pomeau, *Soft Matter*, 2011, 7, 10612-10619.

[17] A. Chakraborti and M. K. Chaudhury, *Langmuir*, 2013, 29, 6926-6935.

[18] D. L. Henann and K. Bertoldi, *Soft Matter*, 2014, 10, 709-717.

[19] R. W. Style and E. R. Dufresne, *Soft Matter*, 2012, 8, 7177-7184.

[20] R. W. Style, Y. Che, J. S. Wettlaufer, L. A. Wilen and E. R. Dufresne, *Phys. Rev. Lett.*, 2013, 110, 066103.

[21] R. W. Style, Y. Che, S. J. Park, B. M. Weon, J. H. Je, C. Hyland, G. K. German, M. P. Power, L. A. Wilen, J. S. Wettlaufer and E. R. Dufresne, *Proc. Natl. Acad. Sci. U. S. A.*, 2013, 110, 12541-12544.

[22] N. Nadermann, C.-Y. Hui and A. Jagota, *Proc. Natl. Acad. Sci. U. S. A.*, 2013, 110, 10541-10545.

[23] J. B. Bostwick, M. Shearer and K. E. Daniels, *Soft Matter*, 2014, 10, 7361-7369.

[24] S. Karpitschka, S. Das, M. van Gorcum, H. Perrin, B. Andreotti and J. H. Snoeijer, *Nat. Commun.*, 2015, 6:7891, DOI: 10.1038/ncomms8891.

[25] R. W. Style, C. Hyland, R. Boltynskiy, J. S. Wettlaufer and E. R. Dufresne, *Nat. Commun.*, 2013, 4:2728, DOI: 10.1038/ncomms3728.

[26] T. Salez, M. Benzaquen and E. Raphael, *Soft Matter*, 2013, 9, 10699-10704.

[27] X. Xu, A. Jagota and C.-Y. Hui, *Soft Matter*, 2014, 10, 4625-4632.

[28] Z. Cao, M. J. Stevens and A. V. Dobrynin, *Macromolecules*, 2014, 47, 3203-3209.

[29] S. Mora, C. Maurini, T. Phou, J.-M. Fromental, B. Audoly and Y. Pomeau, *Phys. Rev. Lett.*, 2013, 111, 114301.

[30] H. L. Duan, X. Yi, Z. P. Huang and J. Wang, *Mech. Mater.*, 2007, 39, 81-93.

[31] S. Brisard, L. Dormieux and D. Kondo, *Comput. Mater. Sci.*, 2010, 48, 589-596.

[32] S. Brisard, L. Dormieux, D. Kondo, *Comput. Mater. Sci.*, 2010, 50, 403-410.

[33] H. L. Duan, J. Wang, Z. P. Huang and B. L. Karihaloo, *Proc. R. Soc. A.*, 2005, 461, 3335-3353.

[34] H. L. Duan, X. Yi, Z. P. Huang and J. Wang, *Mech. Mater.*, 2007, 39, 94-103.

[35] R. W. Style, J. S. Wettlaufer and E. R. Dufresne, *Soft Matter*, 2015, 11, 672-679.

[36] R. W. Style, R. Boltynskiy, B. Allen, K. E. Jensen, H. P. Foote, J. S. Wettlaufer and E. R. Dufresne, *Nature Physics*, 2015, 11, 82-87.

[37] F. Mancarella, R. W. Style, J. S. Wettlaufer, *Interfacial tension and the Mori-Tanaka theory of non-dilute soft composite solids*, subjudice (2015).

[38] H. L. Duan, J. Wang, Z. P. Huang and B. L. Karihaloo, *J. Mech. Phys. Solids*, 2005, 53, 1574-1596.

[39] A.I. Lur’e, *Three-dimensional problems of the theory of elasticity* (translated from the Russian), Interscience, New York, 1964, ch.6, pp. 325-379.

[40] J. D. Eshelby, *Solid State Physics*, 1956, 3, 79-144.

[41] J. D. Eshelby, *Proc. R. Soc. Lond. A*, 1957, 241, 376-396.

[42] J. Dumais and Y. Forterre, *Annual Review of Fluid Mechanics*, 2012, 44, 453-478.

[43] E. Moeendarbary, L. Valon, M. Fritzsche, A. R. Harris, D. A. Moulding, A. J. Thrasher, E. Stride, L. Mahadevan and G. T. Charras, *J. Res. Nat. Bur. Stand. A*, 2012, 117, 947-967.

[44] M. A. Hesse, A. R. Schiemenz, Y. Liang and E. M. Parmenier, *Geophysical Journal International*, 2011, 187, 1057-1075.

[45] M. L. Szulczewski, C. W. MacMinn, H. J. Herzog and R. Juanes, *Proc. Natl. Acad. Sci. U. S. A.*, 2012, 109, 5185-5189.

[46] H. F. Wang, *Theory of Linear Poroelasticity*, Princeton University Press, Princeton NJ, 2000.

[47] C. W. MacMinn, E. R. Dufresne and J. S. Wettlaufer, *Phys. Rev. X*, 2015, 5, 011020.

[48] J. C. Smith, *J. Res. Nat. Bur. Stand. A*, 1974, 78, 355-361.

[49] J. C. Smith, *J. Res. Nat. Bur. Stand. A*, 1975, 79, 419-423.

[50] Note that we could well have chosen \( \varepsilon_0 = (l - 2R)/2R \) rather than \( (l - 2R)/R \), but chose the latter to facilitate comparison to the dilute results from [35].