Stiffening solids with liquid inclusions

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From bone and wood to concrete and carbon fibre, composites are ubiquitous natural and synthetic materials. Eshelby's inclusion theory describes how macroscopic stress fields couple to isolated microscopic inclusions, allowing prediction of a composite's bulk mechanical properties from a knowledge of its microstructure. It has been extended to describe a wide variety of phenomena from solid fracture to cell adhesion. Here, we show experimentally and theoretically that Eshelby's theory breaks down for small liquid inclusions in a soft solid. In this limit, an isolated droplet's deformation is strongly size-dependent, with the smallest droplets mimicking the behaviour of solid inclusions. Furthermore, in opposition to the predictions of conventional composite theory, we find that finite concentrations of small liquid inclusions enhance the stiffness of soft solids. A straightforward extension of Eshelby's theory, accounting for the surface tension of the solid-liquid interface, explains our experimental observations. The counterintuitive stiffening of solids by fluid inclusions is expected whenever inclusion radii are smaller than an elastocapillary length, given by the ratio of the surface tension to Young's modulus of the solid matrix. These results suggest that surface tension can be a simple and effective mechanism to cloak the far-field elastic signature of inclusions.

omposite materials can offer marked performance improvements over their individual components. Carbon fibre increases the strength and stiffness of polymer resins as much as a hundredfold¹. Densely packed gas bubbles in a liquid matrix create a foam, which resists deformation like a solid². The foundational theory of solid composites, due to Eshelby, describes how isolated inclusions in a composite behave in response to applied stresses³. Eshelby applied this result to predict the stiffness of dilute solid composites³ and his theory has been extended to finite concentrations, where neighbouring inclusions couple through their induced strain fields (for example, refs 4,5). Eshelby's theory has been applied widely beyond composites, having long been used to understand the mechanics of fracture^{6,7} and plasticity^{8,9}. More recently, it has been applied to understanding flow of sheared glasses¹⁰ and the interactions of cells with the extracellular matrix^{11,12}.

Eshelby's theory describes the matrix and inclusion as bulk linear-elastic solids, but does not account for the physics of the interface, which generically includes excess surface free energy and surface stress¹³⁻¹⁵. Surface energy is the reversible work per unit area required to create new interfacial area by cutting. Surface stress is the reversible work per unit area to create new interfacial area by stretching. For liquids, surface energy and surface stress are identical, isotropic and strain-independent. For solids, surface stress and energy are generally anisotropic and distinct, but can be isotropic for soft amorphous solids such as gels^{16,17}. Cell membranes and other thin-walled vessels can exhibit large isotropic surface stress with negligible surface energy^{18,19}. In this manuscript, we use the phrase surface tension, denoted by Υ , to denote an isotropic strain-independent surface stress.

Recent work has underlined the importance of surface-tension effects in soft solids. These solid capillary effects include the smoothing out of ripples and corners in soft solids²⁰, and qualitative changes to the phenomena of wetting²¹⁻²⁶ and adhesion²⁷⁻³⁰. Furthermore, the competition of surface tension and elasticity

can select the wavelength of pearling and creasing instabilities^{31,32}. Surface-tension effects typically appear in solids at length scales $\lesssim L \equiv \Upsilon/E$, where *E* is Young's modulus of the solid. In simple terms, this elastocapillary length represents the wavelength below which surface tension is capable of significantly deforming a solid^{21,27}. Thus, it is reasonable to expect that when inclusions in an elastic body have a characteristic size R < L, capillarity will become important and Eshelby's theory will not apply. This has been suggested by various theoretical studies (for example, refs 33,34) and recent experiments on air bubbles embedded in emulsions³⁵.

Here, we demonstrate the impact of surface tension on the mechanical response of fluid inclusions in a soft solid matrix. We find that the deformation of isolated liquid inclusions in a macroscopic stress field depends strongly on their size. Although large-droplet deformations are consistent with Eshelby theory, droplets with radii below the elastocapillary scale deform significantly less than predicted. Furthermore, whereas finite concentrations of large droplets make a solid more compliant, droplets smaller than the elastocapillary scale make it stiffer. A generalization of Eshelby's theory, accounting for surface tension, captures our experimental observations, and provides simple analytical results useful for the design of composites.

Stretching single inclusions

We tested Eshelby's inclusion theory in soft solids by observing the microscopic deformation of droplets embedded in macroscopically deformed solids (Fig. 1a,b and Supplementary Section 1). We coated the soft solid on a thin, elastic sheet, and stretched it uniaxially, measuring the exact applied strain $(\epsilon_x^{\infty}, \epsilon_y^{\infty})$ by tracking fluorescent particles attached to the surface of the sheet (Supplementary Fig. 1). The applied strain in the uniaxial stretch direction is ϵ_x^{∞} and ϵ_y^{∞} is the smaller, associated contraction that arises in the perpendicular, in-plane direction. The stretch lengthens the droplets in the *x*-direction, and we imaged them at their equator from below with a ×60, NA 1.2, water objective. The droplets are ionic

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Figure 1 | **Stretching droplets embedded in soft solids. a**, The sample is clamped and stretched in the *x*-direction. **b**, Example images of ionic-liquid droplets in a soft, silicone solid E = 1.7 kPa. Larger droplets deform more at the same applied strain. Overlay shows small (blue), medium (red) and large (green) droplet images combined together for shape comparison.

liquid (1-ethyl-3-methylimidazolium dicyanamide, Ionic Liquids Technologies) and are completely immiscible in the silicone gel that we use for the solid phase. Silicone gels of two different stiffnesses (E = 1.7 kPa, E = 100 kPa) were prepared by mixing together base and crosslinker at different ratios and curing at room temperature for 16 h, as described in Supplementary Section 2. Silicone gel is ideal for these experiments as it behaves like a linear-elastic solid up to large strains. Supplementary Fig. 2 shows example rheology for the soft, E = 1.7 kPa silicone.

The theory of Eshelby predicts that stretched inclusion shapes depend only on the applied strain, and not on droplet size. We confirmed this result for droplets embedded in a stiff, 100 kPa matrix (Supplementary Fig. 3). However, micrometre-sized droplets behave quite differently in a compliant 1.7 kPa matrix (Fig. 1b). Here, small droplets are significantly less deformed than large droplets under the same macroscopic strain.

Small liquid droplets seems stiffer than the surrounding solid matrix. Figure 2a gives the aspect ratio, $AR = \ell/w$, of eight droplets of different initial radii R at different stretches. As expected for a linear-elastic solid, the aspect ratio increases linearly with ϵ_x^{∞} . However, it also increases with R. In other words, large droplets are deformed more by the stretch, and smaller droplets seem 'stiffer'. The dotted/continuous line shows Eshelby's predictions: $AR = (3+5\epsilon_x^{\infty})/(3+5\epsilon_y^{\infty})$ for incompressible, spherical liquid inclusions, and AR = 1 for rigid spherical inclusions³. We also plot Eshelby's prediction for an inclusion identical to the surrounding solid, $AR = (1 + \epsilon_x^{\infty})/(1 + \epsilon_y^{\infty})$ as the dashed-dotted line—this represents the bulk deformation of the solid matrix. The largest droplet agrees well with the incompressible liquid limit. Smaller droplets seem stiffer, with the smallest droplets approaching the rigid inclusion limit.

The stiffening effect in small droplets seems to arise at a strainindependent length scale. Figure 2b shows the aspect ratio of many droplets as a function of their length, ℓ , for six different strains. For each strain, the aspect ratio is insensitive to the size of large droplets ($\gtrsim 30 \,\mu$ m). However, *AR* drops off sharply for smaller droplets. It is interesting to note that a qualitatively similar dependence of shape on size is seen for droplets in viscous shear or extensional flows³⁶.

Composite stiffness

According to Eshelby's classic result³, liquid inclusions, which have zero Young's modulus, should reduce the stiffness of a solid composite. However, our data show that small, isolated droplets resist deformation more strongly than one would expect from Eshelby's theory. Now, we explore the impact of the increase in apparent stiffness of single droplets on the macroscopic stiffness of a composite. We made soft composites out of silicone gel and glycerol droplets by mixing silicone, glycerol (Sigma-Aldrich) and a small quantity of surfactant (Gransurf 50C-HM, Grant Industries) using a hand blender. Glycerol is used in place of the ionic liquid as it is cheap, non-toxic, and almost completely immiscible in silicone. We degassed the resulting emulsion in a vacuum, poured it into a mould and then cured it at 60 °C for two hours. This gives composites of droplets embedded in silicone with $R = O(1 \,\mu\text{m})$, at volume fractions ϕ from 4 to 20% (Supplementary Fig. 4). As explained in Supplementary Section 2b and Fig. 5, we ignore composites with $\phi < 4.4\%$ to ensure that the stiffness of the continuous phase of the composite is unaffected by the surfactant.

Stiff and compliant solids have opposing responses to liquid inclusions. We measured the composite Young's modulus E_c by macroscopic indentation (see Supplementary Section 2 for detailed protocols). Figure 3a,b shows how composite stiffness changes with increasing liquid content for composites with a stiffer solid matrix with $E \sim 100$ kPa and a more compliant solid matrix with $E \sim 3$ kPa, respectively. The stiff-matrix composite becomes softer as the liquid content increases. This makes intuitive sense-as we replace a fraction of the solid by holes with no shear modulus, we see a proportional decrease to the stiffness. In fact, the data agrees with Eshelby's prediction for the stiffness of a solid containing dilute embedded monodisperse, incompressible droplets, $E_c = E/(1 + 5\phi/3)$ (ref. 3). The compliant-matrix composite shows the opposite trend: stiffening with liquid content. Stiffness increases by around a third with a 20% increase in liquid content. The composites are elastic up to shear strains of ~100%, and behave identically in subsequent cycles of indentation (see Supplementary Section 2c and Fig. 6). Thus, we find that the soft-matrix composite is unexpectedly stiffer than the pure soft solid, without a significant loss in strength. Conventional composite theory, such as Eshelby theory³, the law of mixtures, and the Hashin-Shtrikman bounds⁴ uniformly predict decreasing stiffness with increased fraction of liquid inclusions and, therefore, cannot describe this behaviour (for example, Fig. 3). In a similar vein, recent experiments have shown that the stiffness of an emulsion was unaffected when embedded bubbles were sufficiently small³⁵.

Theory and discussion

The experimental data suggest that conventional composite theory fails to describe our experiments because of the effect of surface tension at the liquid/solid interface. Surface tension typically acts to smooth out interfaces and drive them towards a constant curvature. In a solid, surface tension is typically overwhelmed by bulk elasticity. However, surface tension can cause significant deformations in compliant solids (for example, ref. 37). In our experiments, surface tension acts to keep liquid inclusions spherical, opposing any applied stretch. Thus, surface tension can qualitatively explain the main features of our data. This echoes recent results

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Figure 2 | Aspect ratio of stretched ionic-liquid droplets in a soft (*E*=1.7 kPa) silicone gel as a function of size and strain. **a**, Aspect ratios of eight droplets of different sizes increase linearly with applied strain. The dashed-dotted line shows the change in aspect ratio of the solid under the applied strain ϵ_x^{∞} , ϵ_y^{∞} . Large droplets ($R \gtrsim 5 \,\mu$ m) stretch more than the solid. Smaller droplets stretch less. The dotted (continuous) line shows the prediction from Eshelby theory for incompressible liquid (rigid) solid inclusions respectively³. **b**, Aspect ratio of droplets depends sensitively on size. Different colours correspond to different applied strains. Dashed curves show theoretical predictions using equations (2) and (3) with $\Upsilon = 0.0036 \,\mathrm{N m^{-1}}$.

on wetting and adhesion on compliant silicones, where capillary affects arose below a length scale of $O(10 \,\mu\text{m})$, similar to that seen in Fig. $2b^{22,23,27}$. Here, we modify Eshelby theory to account for solid surface tension, and show that it accurately describes our data.

We consider an incompressible droplet embedded in a linearelastic solid with a surface tension that acts on the droplet boundary. The solid's displacements, **u**, obey the equation:

$$(1-2\nu)\nabla^2 \mathbf{u} + \nabla(\nabla \cdot \mathbf{u}) = 0 \tag{1}$$

where ν is Poisson's ratio and we apply far-field strain boundary conditions $\epsilon = \epsilon^{\infty}$. At the surface of the droplet, $\sigma \cdot \mathbf{n} = -p\mathbf{n} + \Upsilon \mathcal{K}\mathbf{n}$, where σ is the stress tensor in the solid, \mathbf{n} is the normal vector to the deformed surface, p is the pressure in the droplet and \mathcal{K} is the curvature of the deformed surface. Note that we assume a surface tension that is independent of surface strain. This is generally a good approximation for gels, although it is not true in general^{17,34}. We derive analytic solutions to equation (1) (ref. 38) by extending previous work³⁹. In the particular case of far-field, plane-stress



Figure 3 | Young's modulus of soft composites as a function of liquid content. Glycerol droplets embedded in $E \sim 100$ kPa (**a**) and $E \sim 3$ kPa (**b**) silicone gels. Dashed curves show Eshelby's predictions for incompressible liquid droplets in an incompressible solid with Young's modulus *E*.

boundary conditions (as in our experiment) $\epsilon_{xx} = \epsilon_x^{\infty}$, $\epsilon_{yy} = \epsilon_y^{\infty}$ and $\sigma_{zz} = 0$, the length and width of the stretched droplet are

$$\ell = 2R \left[1 + \frac{5(2\epsilon_1 - \epsilon_2)}{6 + 15\frac{\gamma}{ER}} \right]$$
(2)

and

$$w = 2R \left[1 + \frac{5(2\epsilon_2 - \epsilon_1)}{6 + 15\frac{\gamma}{ER}} \right]$$
(3)

where $\epsilon_1 = (\epsilon_x^{\infty} + \nu \epsilon_y^{\infty})/(1 - \nu^2)$ and $\epsilon_2 = (\nu \epsilon_x^{\infty} + \epsilon_y^{\infty})/(1 - \nu^2)$. In the limit $\Upsilon = 0$, this reduces to Eshelby's predictions. In the limit $\Upsilon/ER \gg 1$, surface tension dominates and the droplets stay spherical, as the elastic stresses become insufficient to deform the droplet from its preferred shape. The dependence on the parameter Υ/ER indicates that surface-tension effects start to arise when the size of the droplets approaches the elastocapillary length $L = \Upsilon/E$ (ref. 40). This is similar to previous experiments where solid capillarity becomes important: for example, contact mechanics results are altered when the size of the indenter is $\lesssim L$ (refs 27–29), droplet contact angles change when drop radii are $\lesssim L$ (refs 21,22), and thin fibres undergo instabilities when their diameters are $\lesssim L$ (ref. 31).

Our theory agrees well with the isolated droplet data with one fitting parameter—the unknown surface tension Υ . In Fig. 2b, we plot the aspect ratio predicted by equations (2) and (3), using $\nu = 1/2$, E = 1.7 kPa and $\Upsilon = 0.0036$ N m⁻¹. The results agree with the experiments up to large strains, suggesting that surface tension is indeed controlling droplet shape for small droplets. The agreement is surprisingly good as we use a linear-elastic theory which is only strictly appropriate when $\epsilon \lesssim 10\%$, or equivalently when ϵ_x^{∞} , $\epsilon_y^{\infty} \lesssim 10\%$ (ref. 38). Note that the value of the surface tension is smaller than we expected; we measured surface tension of an ionic

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liquid in uncured silicone to be 0.025 N m^{-1} using the pendant drop method, and we might expect this value to be close to Υ . This difference cannot be explained by measurement error, suggesting that there is a significant change in the silicone/ionic-liquid interface on crosslinking. This is not unprecedented—previous measurements have shown that there can be significant differences between liquid and solid surface tensions of silicone^{22,24,41}.

The individual droplet data collapses onto a single master curve when the ratio of the microscopic to macroscopic strains is plotted against the undeformed droplet radius (Fig. 4a). Using equations (2) and (3), we can obtain an estimate of the undeformed radii, R^* (*R* is unknown, as we did not track individual droplets from their undeformed state for this large data set):

$$R^* = \frac{\ell}{2} - \frac{A}{2}(2\epsilon_1 - \epsilon_2) \tag{4}$$

where

$$A \equiv \frac{(\ell - w)(1 + \nu)}{3(\epsilon_x^{\infty} - \epsilon_y^{\infty})} = \frac{10ER^2}{6ER + 15\Upsilon}$$
(5)

and the second equality comes from equations (2) and (3). The ratio $2A/R^*$ compares the microscopic droplet strain to the macroscopic applied strain. Moreover, $2A/R^*$ and R^* depend only on measured quantities, and nicely collapse the data over a factor of 70 in droplet size, and a range of strains from 5.6 to 42.2% (Fig. 4a). There are two regimes: for droplets of size $R^* \leq 10 \,\mu\text{m}, \,(\ell - w)/R^* \propto (\epsilon_x^\infty - \epsilon_y^\infty)R^*$, whereas for larger droplets $(\ell - w)/R^* \propto (\epsilon_x^\infty - \epsilon_y^\infty)R^*$, whereas for larger droplets as the crossover from capillary- to elastic-dominated regimes as R crosses Υ/E . Note that although our theory effectively collapses the data onto a universal curve, the data in the capillary regime seems to have a stronger dependence on droplet size than predicted.

Our isolated droplet theory can be applied to predict composite stiffnesses^{3,42}. Eshelby showed that the stiffness of a composite consisting of identical dilute inclusions can be calculated from the excess energy of individual strained inclusions³; if the extra strain energy due to the presence of a single inclusion in a uniaxially stretched solid is $W(\sigma^{\infty}, E, R, \Upsilon)$, where σ^{∞} is the applied stress, then the average strain energy density in a dilute composite is

$$\mathcal{E} = \frac{1}{2} \frac{(\sigma^{\infty})^2}{E} + \frac{\phi W}{\frac{4}{3}\pi R^3}$$
(6)

and Young's modulus of the composite is $E_c = (\sigma^{\infty})^2/2\mathcal{E}$. We can use equation (6) to predict the stiffness of a composite with monodisperse, incompressible inclusions with surface tension³⁸. For the particular case of an incompressible solid,

$$E_{c} = E \frac{1 + \frac{5}{2} \frac{\gamma}{ER}}{\frac{5}{2} \frac{\gamma}{ER} (1 - \phi) + (1 + \frac{5}{3}\phi)}$$
(7)

In the limit of small surface tension, or large droplets $(R \gg \Upsilon/E)$, this reduces to Eshelby's result for liquid droplets in an elastic solid, which is $E_c = E/(1 + 5\phi/3)$. When elasticity dominates over surface tension $(R \gg \Upsilon/E)$, we obtain $E_c = E/(1 - \phi)$, and the material is stiffened by the inclusions. This differs from Eshelby's result for rigid particles embedded in an elastic composite, $E_c = E/(1 - 5\phi/2)$ although surface tension keeps the droplets spherical, they are distinct from rigid particles because they have zero shear stress at their surfaces. Equation (7) predicts that composites are stiffened by droplets when $R < 1.5\Upsilon/E$. Figure 4b shows how composite stiffness depends on liquid fraction, as predicted by equation (7) for different values of Υ/ER . Intriguingly, we predict no change of the effective modulus of the composite when $R = 1.5\Upsilon/E$. This suggests that



Figure 4 | Theoretical predictions of composite behaviour. a, The data from Fig. 2b collapses onto a master curve when plotting the ratio of the microscopic to macroscopic strains (equations (4) and (5)) against the estimated radius (equation (4)). The dashed curve shows the theoretical prediction in equation (5). b, Effective Young's modulus of a composite consisting of monodisperse droplets embedded in a uniform solid, from equation (7). The blue(black) data points are the composite stiffness data from Fig. 3 scaled by E = 3 kPa (E = 100 kPa) for the soft (stiff) composites respectively. **c**, Droplets with surface tension can be considered as equivalent elastic inclusions without surface tension. The red curve shows how the stiffness of the equivalent elastic inclusion, E_i , depends on Υ/ER . For small Υ/ER , this agrees fairly well with the approximation $E_i = 2\Upsilon/R$, shown by the blue line.

surface tension can effectively cloak the far-field elastic signature of inclusions^{43,44}.

This theory for composite stiffness is consistent with our experimental data. Figure 4b includes the data from Fig. 3, normalized by E = 3 kPa and E = 100 kPa for the softer and stiffer composites respectively. The soft-matrix composite results are modelled well by the surface-tension-dominated theory. The stiff-matrix composite results are modelled well by the theory with little, or no, surface-tension effects. Using rough estimates



of $\Upsilon \sim 14 \text{ mN m}^{-1}$ (Supplementary Section 2b) and $R \sim 1 \,\mu\text{m}$, we indeed expect surface tension to dominate for the soft-matrix composite ($\Upsilon/ER > 1$), and to be small for the stiff-matrix composite ($\Upsilon/ER < 1$). Note that the experimental data for the soft-matrix composite is consistently stiffer than the upper limit of our theory. We suspect that this is due to formation of chain-like structures of droplets (Supplementary Fig. 4). Our theory is strictly valid only in the limit of isolated droplets.

We can greatly simplify the above results to give a simple physical picture of the effect of surface tension in soft composites. The stiffness of a composite of incompressible elastic inclusions with Young's modulus E_i in a solid of modulus E, according to Eshelby³, is

$$E_{c} = E \frac{1 + \frac{2}{3} \frac{E_{i}}{E}}{\left(\frac{2}{3} - \frac{5\phi}{3}\right) \frac{E_{i}}{E} + \left(1 + \frac{5}{3}\phi\right)}$$
(8)

If we equate equations (7) and (8), we find that embedded droplets are equivalent to elastic inclusions⁴² with stiffness

$$E_{i} = E \frac{24 \frac{\gamma}{ER}}{10 + 9 \frac{\gamma}{ER}}$$

$$\tag{9}$$

This recovers the result of ref. 35 (derived using equivalent inclusion dipoles) that was successfully used to describe bubbles in soft emulsions. When $\gamma/ER \ll 1$, the droplets behave like inclusions with Young's modulus $E_i = 12\gamma/5R$. This value is close to the droplet Laplace pressure $E_i = 2\Upsilon/R$, which is often taken as its stiffness for describing the composite stiffness of emulsions and gels⁴⁵. In the capillary-dominated regime, $\gamma/ER \gg 1$, the effective Young's modulus of the inclusions saturates at $E_i = 8E/3$. Thus the droplets cannot have an arbitrarily increasing effective stiffness as they get smaller, as the common $E_i = 2\Upsilon/R$ ansatz suggests. By replacing capillary-dominated inclusions with equivalent elastic inclusions described by equation (9) (for example, refs 35,42), one can use established composite theory such as Mori-Tanaka homogenization^{5,35} or self-consistent methods⁴⁶ to predict denser composite stiffnesses. Future work can also generalize the Hashin-Shtrikman bounds on composite moduli⁴ to include surface tension-either using equivalent elastic inclusions, or by using the thin-layer analogy for interface area introduced for the case of surface-strain-dependent surface stresses^{15,47}.

Our experimental and theoretical results show that surface tension can be important for soft composites consisting of a liquid phase embedded in a continuous solid phase. We expect that surface tension will be important for solid/solid composites whenever $R \leq 100\Upsilon/E_1$, $100\Upsilon/E_2$, where E_1 , E_2 are the stiffnesses of the two solids. For compliant materials such as gels with E = O(kPa), capillarity needs to be addressed at scales of up to $O(100 \,\mu\text{m})$ (refs 31,35). For stiffer materials, such as elastomers, with E = O(MPa), capillarity needs to be addressed at scales of up to $O(100 \,\mu\text{m})$. Capillary effects should negligible in structural materials, such as glass and ceramics, with E = O(GPa).

We expect that our results should be of use in understanding the mechanical properties of soft tissues, especially in soft connective tissues. For example, the cortical tension of fibroblasts may have a larger impact on the bulk mechanical properties of a collagenous tissue than the fibroblasts' elastic moduli^{48,49}. Our results complement new approaches to measuring mechanical forces within three dimensional tissues by quantification of the deformation of embedded liquid droplets⁵⁰.

Our theoretical results include simple analytic expressions for individual droplet deformation and for the properties of the bulk composite that can be readily applied to the design of new materials. They suggest that surface tension provides a relatively simple and effective means to cloak the elastic signature of inclusions in soft materials. Received 15 July 2014; accepted 10 November 2014; published online 15 December 2014

References

- Ashby, M. F. Overview no. 80: On the engineering properties of materials. *Acta Metall.* 37, 1273–1293 (1989).
- Höhler, R. & Cohen-Addad, S. Rheology of liquid foam. J. Phys. Condens. Matter 17, R1041 (2005).
- 3. Eshelby, J. D. The determination of the elastic field of an ellipsoidal inclusion, and related problems. *Proc. R. Soc. Lond. A* **241**, 376–396 (1957).
- Hashin, Z. & Shtrikman, S. A variational approach to the theory of the elastic behaviour of multiphase materials. *J. Mech. Phys. Solids* 11, 127–140 (1963).
- Mori, T. & Tanaka, K. Average stress in matrix and average elastic energy of materials with misfitting inclusions. *Acta Metall.* 21, 571–574 (1973).
- 6. Rice, J. R. A path independent integral and the approximate analysis of strain concentration by notches and cracks. *J. Appl. Mech.* **35**, 379–386 (1968).
- Budiansky, B. & O'Connell, R. J. Elastic moduli of a cracked solid. Int. J. Solids Struct. 12, 81–97 (1976).
- Hutchinson, J. Elastic-plastic behaviour of polycrystalline metals and composites. Proc. R. Soc. Lond. A 319, 247–272 (1970).
- Berveiller, M. & Zaoui, A. An extension of the self-consistent scheme to plastically-flowing polycrystals. J. Mech. Phys. Solids 26, 325–344 (1978).
- Jensen, K. E., Weitz, D. A. & Spaepen, F. Local shear transformations in deformed and quiescent hard-sphere colloidal glasses. *Phys. Rev. E* **90**, 042305 (2014).
- Zemel, A., Rehfeldt, F., Brown, A. E. X., Discher, D. E. & Safran, S. A. Optimal matrix rigidity for stress-fibre polarization in stem cells. *Nature Phys.* 6, 468–473 (2010).
- 12. Schwarz, U. S. & Safran, S. A. Physics of adherent cells. *Rev. Mod. Phys.* 85, 1327–1381 (2013).
- Gurtin, M. E. & Murdoch, A. I. A continuum theory of elastic material surfaces. Arch. Ration. Mech. Anal. 57, 291–323 (1975).
- 14. Palierne, J. F. Linear rheology of viscoelastic emulsions with interfacial tension. *Rheol. Acta* **29**, 204–214 (1990).
- Brisard, S., Dormieux, L. & Kondo, D. Hashin–Shtrikman bounds on the shear modulus of a nanocomposite with spherical inclusions and interface effects. *Comput. Mater. Sci.* 50, 403–410 (2010).
- Shuttleworth, R. The surface tension of solids. Proc. Phys. Soc. A 63, 444–457 (1950).
- Hui, C-Y. & Jagota, A. Surface tension, surface energy, and chemical potential due to their difference. *Langmuir* 29, 11310–11316 (2013).
- Needham, D. & Hochmuth, R. A sensitive measure of surface stress in the resting neutrophil. *Biophys. J.* 61, 1664–1670 (1992).
- Lecuit, T. & Lenne, P-F. Cell surface mechanics and the control of cell shape, tissue patterns and morphogenesis. *Nature Rev. Mol. Cell Biol.* 8, 633–644 (2007).
- Mora, S. et al. Solid drops: Large capillary deformations of immersed elastic rods. Phys. Rev. Lett. 111, 114301 (2013).
- 21. Style, R. W. & Dufresne, E. R. Static wetting on deformable substrates, from liquids to soft solids. *Soft Matter* **8**, 7177–7184 (2012).
- Style, R. W., Che, Y., Wettlaufer, J. S., Wilen, L. A. & Dufresne, E. R. Universal deformation of soft substrates near a contact line and the direct measurement of solid surface stresses. *Phys. Rev. Lett.* **110**, 066103 (2013).
- Style, R. W. et al. Patterning droplets with durotaxis. Proc. Natl Acad. Sci. USA 110, 12541–12544 (2013).
- Nadermann, N., Hui, C-Y. & Jagota, A. Solid surface tension measured by a liquid drop under a solid film. *Proc. Natl Acad. Sci. USA* 110, 10541–10545 (2013).
- Karpitschka, S., Das, S., Andreotti, B. & Snoeijer, J. Dynamic contact angle of a soft solid. Preprint at http://arXiv.org/abs/1406.5547v1 (2014).
- Bostwick, J. B., Shearer, M. & Daniels, K. E. Elastocapillary deformations on partially-wetting substrates: Rival contact-line models. *Soft Matter* 10, 7361–7369 (2014).
- Style, R. W., Hyland, C., Boltyanskiy, R., Wettlaufer, J. S. & Dufresne, E. R. Surface tension and contact with soft elastic solids. *Nature Commun.* 4, 2728 (2013).
- Salez, T., Benzaquen, M. & Raphael, E. From adhesion to wetting of a soft particle. *Soft Matter* 9, 10699–10704 (2013).
- Xu, X., Jagota, A. & Hui, C-Y. Effects of surface tension on the adhesive contact of a rigid sphere to a compliant substrate. *Soft Matter* 10, 4625–4632 (2014).
- Cao, Z., Stevens, M. J. & Dobrynin, A. V. Adhesion and wetting of nanoparticles on soft surfaces. *Macromolecules* 47, 3203–3209 (2014).
- Mora, S., Phou, T., Fromental, J-M., Pismen, L. M. & Pomeau, Y. Capillarity driven instability of a soft solid. *Phys. Rev. Lett.* 105, 214301 (2010).

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- Chen, D., Cai, S., Suo, Z. & Hayward, R. C. Surface energy as a barrier to creasing of elastomer films: An elastic analogy to classical nucleation. *Phys. Rev. Lett.* 109, 038001 (2012).
- Yang, F. Size-dependent effective modulus of elastic composite materials: Spherical nanocavities at dilute concentrations. *J. Appl. Phys.* 95, 3516–3520 (2004).
- Sharma, P. & Ganti, S. Size-dependent eshelbyös tensor for embedded nano-inclusions incorporating surface/interface energies. J. Appl. Mech. 71, 663–671 (2004).
- Ducloue, L., Pitois, O., Goyon, J., Chateau, X. & Ovarlez, G. Coupling of elasticity to capillarity in soft aerated materials. *Soft Matter* 10, 5093–5098 (2014).
- 36. Cox, R. G. The deformation of a drop in a general time-dependent fluid flow. *J. Fluid Mech.* **37**, 601–623 (1969).
- Chakrabarti, A. & Chaudhury, M. K. Direct measurement of the surface tension of a soft elastic hydrogel: Exploration of elastocapillary instability in adhesion. *Langmuir* 29, 6926–6935 (2013).
- Style, R. W., Wettlaufer, J. S. & Dufresne, E. R. Surface tension and the mechanics of liquid inclusions in compliant solids. *Soft Matter* (in the press). Preprint at http://arXiv.org/abs/1409.1998 (2014).
- Duan, H. L., Wang, J., Huang, Z. P. & Karihaloo, B. L. Eshelby formalism for nano-inhomogeneities. *Proc. R. Soc. A* 461, 3335–3353 (2005).
- Wang, J., Duan, H., Huang, Z. & Karihaloo, B. A scaling law for properties of nano-structured materials. *Proc. R. Soc. A* 462, 1355–1363 (2006).
- 41. Park, S. J. *et al.* Visualization of asymmetric wetting ridges on soft solids with X-ray microscopy. *Nature Commun.* **5**, 4369 (2014).
- Duan, H. L., Yi, X., Huang, Z. P. & Wang, J. A unified scheme for prediction of effective moduli of multiphase composites with interface effects. part i: Theoretical framework. *Mech. Mater.* **39**, 81–93 (2007).
- Milton, G. W., Briane, M. & Willis, J. R. On cloaking for elasticity and physical equations with a transformation invariant form. *New J. Phys.* 8, 248 (2006).
- 44. Bückmann, T., Thiel, M., Kadic, M., Schittny, R. & Wegener, M. An elasto-mechanical unfeelability cloak made of pentamode metamaterials. *Nature Commun.* **5**, 4130 (2014).
- Dickinson, E. Emulsion gels: The structuring of soft solids with protein-stabilized oil droplets. *Food Hydrocolloids* 28, 224–241 (2012).

- 46. Hill, R. A self-consistent mechanics of composite materials. J. Mech. Phys. Solids 13, 213–222 (1965).
- Brisard, S., Dormieux, L. & Kondo, D. Hashin–Shtrikman bounds on the bulk modulus of a nanocomposite with spherical inclusions and interface effects. *Comput. Mater. Sci.* 48, 589–596 (2010).
- Brown, R., Prajapati, R., McGrouther, D., Yannas, I. & Eastwood, M. Tensional homeostasis in dermal fibroblasts: Mechanical responses to mechanical loading in three-dimensional substrates. J. Cell. Physiol. 175, 323–332 (1998).
- Zemel, A., Bischofs, I. & Safran, S. Active elasticity of gels with contractile cells. Phys. Rev. Lett. 97, 128103 (2006).
- Campàs, O. et al. Quantifying cell-generated mechanical forces within living embryonic tissues. Nature Methods 11, 183–189 (2014).

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Author contributions

R.W.S., R.B., B.A., K.E.J., H.P.F. and E.R.D. designed experiments. R.W.S., B.A. and R.B. acquired data. R.W.S. and E.R.D. analysed data. R.W.S., J.S.W. and E.R.D. developed the theory. R.W.S., J.S.W. and E.R.D. wrote the paper.

Additional information

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Competing financial interests

The authors declare no competing financial interests.