

Contact angles on a soft solid: from Young's law to Neumann's law (Supplementary Material)

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In this Supplementary Online Material, we describe the technical details of the results provided in the main paper. First, we discuss how to compute capillary forces in the vicinity of the contact line within the Density Functional Theory, in case the solid and the liquid phases can be considered as perfect wedges. In this framework, we determine the equilibrium of a liquid when the solid is not a flat substrate, but a wedge. We show how Young's law and Neumann's law are recovered in particular limits. Second, we present a macroscopic linear model for the elastic response of the solid to capillary forces. This is the basis for the analytical and numerical results presented in the main paper.

CALCULATION OF CAPILLARY FORCES IN WEDGES

The DFT framework in which we perform our analysis separates the molecular interactions into a long-range attractive potential and a short-range hard core repulsion [1, 2]. The short-range repulsion is described by an internal repulsive pressure p_r that ensures incompressibility of both the liquid and the solid – this is a reasonable approximation for elastomers. This pressure acts as a purely normal contact force, and is continuous across the interfaces. Using Eq. 2 of the main text to define Φ_{SL} and Φ_{LL} , the mechanical equilibrium allows one to express p_r in the liquid as (see [2] for the derivation):

$$p_r + \Phi_{SL} + \Phi_{LL} = cst. \quad (1)$$

The force \vec{F}_{LS} exerted by a wedge of liquid on a subsystem of the solid of volume \mathcal{V}_S , can be expressed as the resultant of the volumetric attraction and the repulsion at the liquid-solid interface \mathcal{S}_{LS} . Within the same framework, one can compute the force \vec{F}_{SL} exerted by a wedge of solid on a subsystem of the liquid of volume \mathcal{V}_L . One writes:

$$\vec{F}_{LS} = - \int_{\mathcal{V}_S} \vec{\nabla} \Phi_{LS}(r) d^3r - \int_{\mathcal{S}_{LS}} p_r \vec{n} d^2r, \quad (2)$$

$$\vec{F}_{SL} = - \int_{\mathcal{V}_L} \vec{\nabla} \Phi_{SL}(r) d^3r - \int_{\mathcal{S}_{LS}} p_r \vec{n} d^2r. \quad (3)$$

When the liquid and the solid domains are perfect wedges, these integrals can be evaluated explicitly, and expressed in terms of the surface tensions $\gamma, \gamma_{SL}, \gamma_{SV}$. This will be derived below and lead to the equations of the main paper, in particular Eqs. 3, 5 and 6.

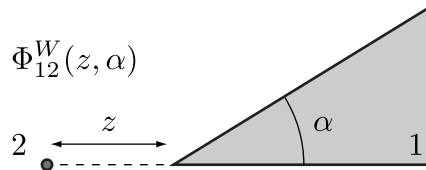


FIG. 1: Definition of the attractive potential $\Phi_{12}^W(z, \alpha)$, created by a wedge (of angle α) of phase 1 on a point of phase 2. The expression is given in (4).

First, we determine the equilibrium shape of a liquid wedge on a given solid wedge, and the force exerted by the liquid on the solid in the vicinity of the contact line. This calculation can be reduced entirely in terms of the interactions between wedges, as in Fig. 1. The attractive potential $\Phi_{12}^W(z, \alpha)$ created by a wedge of a phase 1, and

characterized by an angle $0 < \alpha < \pi/2$, on a point of phase 2, at a distance z from the corner, and aligned with one of the two edges (see Fig. 1) can be analytically expressed as:

$$\Phi_{12}^W(z, \alpha) = \int_z^\infty r \left[\alpha - \arcsin \left(\frac{z}{r} \sin \alpha \right) \right] \tilde{\varphi}_{12}(r) dr. \quad (4)$$

where $\tilde{\varphi}_{12}(r) = \rho_1 \rho_2 \int_{-\infty}^\infty \varphi_{12}(\sqrt{r^2 + y^2}) dy$ is the potential created by an infinite line at a distance r from it. Note that by construction, this function must be even with respect to the angle. The more general case when a point is not aligned with an edge of the wedge can be easily deduced from the above case by adding (or subtracting) two different wedges. Furthermore, as the surface tensions are calculated from the interactions between a column and half a space, we can obtain certain important relations governing $\tilde{\varphi}_{12}(r)$ [3]:

$$\int_0^\infty r^2 \tilde{\varphi}_{LL}(r) dr = -\gamma, \quad (5)$$

$$\int_0^\infty r^2 \tilde{\varphi}_{LS}(r) dr = \int_0^\infty r^2 \tilde{\varphi}_{SL}(r) dr = -\frac{\gamma + \gamma_{SV} - \gamma_{SL}}{2} = -\gamma \frac{1 + \cos \theta_Y}{2}. \quad (6)$$

Here θ_Y is the Young' angle. These two integrals can now be used in (4) to obtain:

$$\int_0^\infty \Phi_{LL}^W(z, \alpha) dz = -\gamma \frac{1 - \cos \alpha}{\sin \alpha}, \quad (7)$$

$$\int_0^\infty \Phi_{LS}^W(z, \alpha) dz = \int_0^\infty \Phi_{SL}^W(z, \alpha) dz = -\gamma \frac{1 + \cos \theta_Y}{2} \frac{1 - \cos \alpha}{\sin \alpha}. \quad (8)$$

The attractive potential $\Phi_{12}^H(z)$ created by a half-space of a phase 1, on a point at a distance z from its flat boundary can be expressed independently or by using two wedges of 90° :

$$\Phi_{12}^H(z) = 2 \int_z^\infty r \arccos \frac{z}{r} \tilde{\varphi}_{12}(r) dr = 2 \Phi_{12}^W \left(z, \frac{\pi}{2} \right). \quad (9)$$

This quantity (noted $\Pi_{12}(z)$ in [2]) can be interpreted as a disjoining pressure. Furthermore, we have some useful symmetry properties that we can easily interpret from geometrical arguments (here we dropped the subscripts to clarify the notations):

$$\Phi^H(z < 0) = 2\Phi^H(0) - \Phi^H(-z), \quad (10)$$

$$\Phi^W(z, \alpha > \pi/2) = \Phi^H(z \sin \alpha) - \Phi^W(z, \pi - \alpha), \quad (11)$$

$$\Phi^W(z < 0, \alpha) = \Phi^H(0) - \Phi^W(-z, \pi - \alpha). \quad (12)$$

Force exerted by the solid on the liquid near the contact line

We denote by \mathcal{S}_{LV} , \mathcal{S}_{SV} and \mathcal{S}_{SL} the liquid-vapor, solid-vapor and solid-liquid interfaces, respectively. The subsystem of liquid subject to forces is bounded on the liquid side by a surface perpendicular to the liquid-solid interface, denoted \mathcal{S}_{LL} (see Fig. 2). The angle of the liquid wedge is $0 < \theta_L < \pi/2$ and the angle of the solid wedge is $0 < \theta_S < \pi$. The force exerted by the solid wedge on the liquid subsystem in the vicinity of the corner is given by (3). Using Ostrogradsky's theorem, the volume integral in (3) reduces to a surface integral, in terms of the surfaces bounding the volume of the liquid subsystem:

$$\vec{F}_{SL} = - \int_{\mathcal{S}_{LV}} \Phi_{SL}^{(1)} (-\sin \theta_L \vec{u}_x + \cos \theta_L \vec{u}_z) dS - \int_{\mathcal{S}_{LL}} \Phi_{SL}^{(2)} \vec{u}_x dS + \int_{\mathcal{S}_{SL}} \left(\Phi_{SL}^{(3)} + p_r^{(3)} \right) \vec{u}_z dS, \quad (13)$$

where \vec{u}_x and \vec{u}_z are the tangential and the normal unit vectors to the solid-liquid interface, both directed toward the interior of the liquid. We thus have to calculate the three different attractive potentials $\Phi_{SL}^{(i)}$ created by the solid wedge on different locations of the liquid wedge, and the repulsive pressure $p_r^{(3)}$ in the liquid at the liquid-solid interface. Let us first concentrate on the liquid at the liquid-solid interface. The liquid is at equilibrium, so we can use Eq. (1) to obtain (in the entire liquid) $p_r + \Phi_{SL} + \Phi_{LL} = \Phi_{LL}^H(0)$, which can be simplified to obtain:

$$p_r^{(3)} + \Phi_{SL}^{(3)} = \Phi_{LL}^W(x, \pi - \theta_L). \quad (14)$$

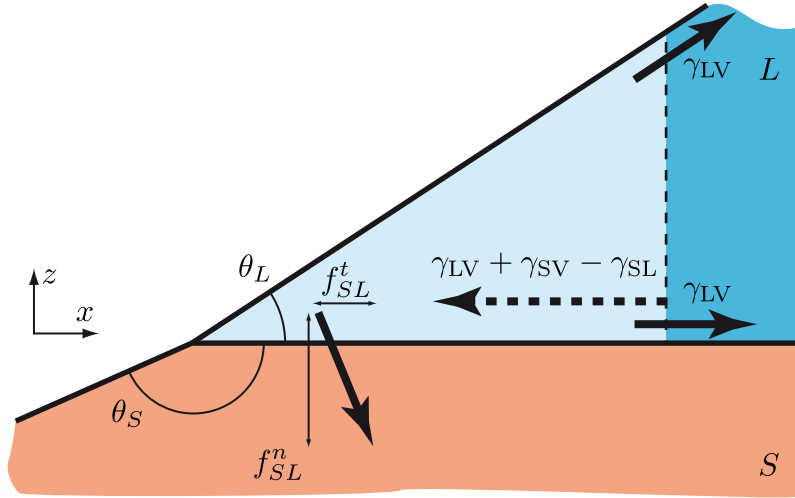


FIG. 2: Distribution of capillary forces induced in a subsystem of the liquid wedge near the contact line.

Then, we can express the solid-liquid attractive potentials $\Phi_{SL}^{(2)}$ and $\Phi_{SL}^{(1)}$ respectively far from the contact line at a distance z from the solid-liquid interface, and along the liquid-vapor interface at a distance s from the contact line:

$$\Phi_{SL}^{(2)} = \Phi_{SL}^H(z), \quad (15)$$

$$\Phi_{SL}^{(1)} = \Phi_{SL}^W(s, \pi - \theta_L) + \Phi_{SL}^W(s, \theta_L + \theta_S - \pi). \quad (16)$$

Using the different expressions of the potentials above (14-16) and the integrals (7,8), we can finally explicitly express the force (13) per unit L of contact line along the two directions normal and tangential to the solid-liquid interface:

$$f_{SL}^t = \frac{\vec{F}_{SL} \cdot \vec{u}_x}{L} = \gamma(1 + \cos \theta_Y) \frac{\sin \theta_L}{2} \left[\frac{1 - \cos \theta_L}{\sin \theta_L} + \frac{1 + \cos(\theta_L + \theta_S)}{\sin(\theta_L + \theta_S)} \right], \quad (17)$$

$$f_{SL}^n = \frac{\vec{F}_{SL} \cdot \vec{u}_z}{L} = \gamma \left[(1 + \cos \theta_Y) \frac{\cos \theta_L}{2} \left(\frac{1 + \cos \theta_L}{\sin \theta_L} - \frac{1 + \cos(\theta_L + \theta_S)}{\sin(\theta_L + \theta_S)} \right) - \frac{1 + \cos \theta_L}{\sin \theta_L} \right]. \quad (18)$$

Liquid equilibrium

As explained in the main text, the equilibrium condition for the liquid is obtained by balancing the solid-on-liquid force with the forces due to liquid-liquid interactions. The system on which the forces act is taken as the liquid corner depicted in Fig. 2). The liquid-liquid interactions can be separated in two contributions. First, there are attractive liquid on liquid forces, which act at the liquid-liquid and the solid-liquid interfaces, far from the contact line. Each of these two forces are equal to γ per unit contact line, and are pulling on these two corners along the liquid-vapor and liquid-solid interfaces. The second is due to repulsive liquid-liquid interactions, acting near the boundary normal to the solid-liquid interface far from the contact line. It originates from the presence of the attractive solid. The resulting tangential force (per length unit of contact line) is $\gamma + \gamma_{SV} - \gamma_{SL} = \gamma(1 + \cos \theta_Y)$ and is directed toward the interior of the subsystem [4]. The balance of these forces, tangential and normal to the interface, read:

$$\gamma(1 + \cos \theta_L) - \gamma(1 + \cos \theta_Y) + f_{SL}^t = 0, \quad (19)$$

$$\gamma \sin \theta_L + f_{SL}^n = 0. \quad (20)$$

These two equations come from the projections of the gradient of a scalar: it implies that they represent a single independent equation that reduces to [using for instance (17) and (19)]:

$$1 + \cos \theta_Y = (1 + \cos \theta_L) \left(1 + \frac{\tan(\theta_L/2)}{\tan(\theta_S/2)} \right). \quad (21)$$

The unique physical solution of this equation is given by:

$$\cos \theta_L = \frac{1}{2} \left[\cos \theta_Y [1 - \cos \theta_S] - \sin \theta_S \sqrt{\frac{2}{1 - \cos \theta_S} - \cos^2 \theta_Y} \right], \quad (22)$$

which is Eq. (3) in the main paper.

Young's law and Neumann's law

One easily shows that in the case of a flat substrate, $\theta_S = \pi$, the tangential force f_{SL}^t vanishes due to symmetry reasons: the solid spans an infinite half-space in the tangential direction. We then recover $\theta_L = \theta_Y$, [either from (19) or (22)], which is Young's law. In addition we note that $f_{SL}^n = \gamma \sin \theta_Y$, ensuring that we have a force balance in the liquid subsystem in both directions.

By assuming the substrate is a perfect liquid, we will now show that the above formulation is equivalent to Neumann's law [5]. Denoting this liquid substrate as phase 2, and the liquid wedge on top of it as phase 1, Neumann's law is given by the two following equations:

$$\gamma_1 \sin \theta_1 - \gamma_2 \sin \theta_2 = 0, \quad (23)$$

$$\gamma_1 \cos \theta_1 + \gamma_2 \cos \theta_2 + \gamma_{12} = 0, \quad (24)$$

where γ_1 and γ_2 are the surface tensions of liquids 1 and 2, γ_{12} is the interfacial tension, and (θ_1, θ_2) are the positive angles made by the liquid wedges 1 and 2. Neumann's law is in fact ruled by two independent parameters, which we denote according to Young's law by:

$$\cos \theta_{Y1} = \frac{\gamma_2 - \gamma_{12}}{\gamma_1}, \quad (25)$$

$$\cos \theta_{Y2} = \frac{\gamma_1 - \gamma_{12}}{\gamma_2}. \quad (26)$$

With these notations, we get $\gamma_1/\gamma_2 = (1 + \cos \theta_{Y2})/(1 + \cos \theta_{Y1})$ and $\gamma_{12}/\gamma_2 = (1 - \cos \theta_{Y1} \cos \theta_{Y2})/(1 + \cos \theta_{Y1})$, and Neumann's law (23,24) can be rewritten as:

$$\sin \theta_1 (1 + \cos \theta_{Y2}) - \sin \theta_2 (1 + \cos \theta_{Y1}) = 0, \quad (27)$$

$$\cos \theta_1 (1 + \cos \theta_{Y2}) + \cos \theta_2 (1 + \cos \theta_{Y1}) = \cos \theta_{Y1} \cos \theta_{Y2} - 1, \quad (28)$$

The unique physical solution can then be explicitly expressed as:

$$\cos \theta_1 = \frac{[2 + (1 - \cos \theta_{Y2}) \cos \theta_{Y1}] \cos \theta_{Y1} - (1 + \cos \theta_{Y2})}{2(1 - \cos \theta_{Y1} \cos \theta_{Y2})}. \quad (29)$$

or equivalently by a function of the form $\theta_1 = N(\theta_{Y1}, \theta_{Y2})$. The second angle is given by symmetry, i.e., $\theta_2 = N(\theta_{Y2}, \theta_{Y1})$.

Let us now show that the DFT formulation indeed reduces to Neumann's law when both the phases are liquid. For a given substrate wedge of angle θ_2 , we can see from (22) that the liquid 1 is at equilibrium when it forms a wedge of angle $\theta_1 = f(\theta_Y, \theta_2)$. If the phase 2 is a liquid as well, then we can use the same equation (22) with exchanged roles of phase 1 and 2. This gives two equations for the contact angles:

$$\theta_1 = f(\theta_{Y1}, \theta_2), \quad (30)$$

$$\theta_2 = f(\theta_{Y2}, \theta_1), \quad (31)$$

where θ_{Y2} is defined in (26). Therefore, θ_1 obeys the following implicit equation:

$$\theta_1 = f(\theta_{Y1}, f(\theta_{Y2}, \theta_1)). \quad (32)$$

We can easily verify that this equation has a unique solution in the interval $[0, \pi]$, which is given explicitly by (29). By analogy, we also recover the second angle θ_2 with the symmetry $\theta_{Y1}|\theta_{Y2}$.

Force exerted by the liquid on the solid near the contact line

Contrarily to the liquid, the capillary forces on the solid need not be at equilibrium for the solid: a resultant force can be balance by the elasticity. By analogy with the determination of the solid on liquid force, we compute the capillary force (2) exerted by the entire wedge of liquid to a subsystem of solid bounded by a surface perpendicular to the liquid-solid interface, noted \mathcal{S}_{SV} :

$$\vec{F}_{LS} = - \int_{\mathcal{S}_{SV}} \Phi_{LS}^{(1)} (-\sin \theta_S \vec{u}_x - \cos \theta_S \vec{u}_z) dS - \int_{\mathcal{S}_{SS}} \Phi_{LS}^{(2)} \vec{u}_x dS - \int_{\mathcal{S}_{SL}} (\Phi_{LS}^{(3)} + p_r^{(3)}) \vec{u}_z dS . \quad (33)$$

The attractive potentials $\Phi_{LS}^{(1)} = \Phi_{LS}^W(s, \pi - \theta_S) + \Phi_{LS}^W(s, \theta_L + \theta_S - \pi)$ and $\Phi_{LS}^{(2)} = \Phi_{LS}^H(z)$ are analogous to what we obtained in (16) and (15). The only fundamental difference is that for this case the pressure is determine by the liquid (and not the solid) equilibrium (14), so that:

$$p_r^{(3)} + \Phi_{LS}^{(3)} = \Phi_{LL}^W(x, \pi - \theta_L) + \Phi_{SL}^W(x, \pi - \theta_S) - \Phi_{LS}^W(x, \pi - \theta_L) . \quad (34)$$

Therefore, after some simplifications, we can determine [from (33)] the normal and the tangential components of the liquid on solid force (per unit length of the contact line):

$$f_{LS}^t = \frac{\vec{F}_{LS} \cdot \vec{u}_x}{L} = \gamma \frac{1 + \cos \theta_Y}{2} \left[1 - \cos \theta_S + \sin \theta_S \frac{1 + \cos(\theta_L + \theta_S)}{\sin(\theta_L + \theta_S)} \right] , \quad (35)$$

$$f_{LS}^n = \frac{\vec{F}_{LS} \cdot \vec{u}_z}{L} = \gamma \left[\frac{1 + \cos \theta_Y}{2} \left(\sin \theta_S + \cos \theta_S \frac{1 + \cos(\theta_L + \theta_S)}{\sin(\theta_L + \theta_S)} \right) + \frac{1 - \cos \theta_Y}{2} \frac{1 + \cos \theta_L}{\sin \theta_L} \right] . \quad (36)$$

These expressions can be rewritten in a more compact form as Eqs. (5) and (6) of the main paper. When $\theta_S = \pi$, then $\theta_L = \theta_Y$ [using (22)] and $f_{LS}^n = \gamma \sin \theta_Y$ [using (36)], as we would expect from the vertical action-reaction interaction of two full wedges, and $f_{LS}^t = \gamma(1 + \cos \theta_Y)$ [using (35)], as was shown in [2, 6].

MACROSCOPIC ELASTO-CAPILLARY MODEL

The description of the solid is more subtle than that of the liquid. The bulk of the elastic substrate is described as a homogeneous incompressible elastic medium. However, one needs to define the effective boundary conditions to be applied to the bulk elasticity equations to compute the deformations. As discussed in the main text, one can consider two models that will be referred to as *normal force transmission* and *vectorial force transmission* respectively. Below we present the expressions for the surface stress σ_s used for our numerical results.

Curved interfaces interactions

For later reference, we first determine the attractive potential at an interface between two phases 1 and 2 which presents a curvature κ . For $\kappa a \ll 1$, this potential (Eq. 2 in the main text) reduces to:

$$\Phi_{12} = \Phi_{12}^H(0) - \kappa \int_0^\infty r^2 \tilde{\varphi}_{12}(r) dr , \quad (37)$$

where, κ is defined as positive when phase 1 is smaller than phase 2. In (37), the integral term is always negative, and depending on whether one considers liquid-liquid, solid-solid or liquid-solid interaction, becomes equal to $-\gamma$, $-\gamma_{SV}$ or $-\gamma(1 + \cos \theta_Y)/2$ [see (7,8)]. The Laplace pressure originates from this potential.

Normal force transmission model

An elastomer can be described as an incompressible soft solid within which the elastic stress can be written as:

$$\sigma_{ij} = -p_r \delta_{ij} + \frac{E}{3} (\partial_i u_j + \partial_j u_i) , \quad (38)$$

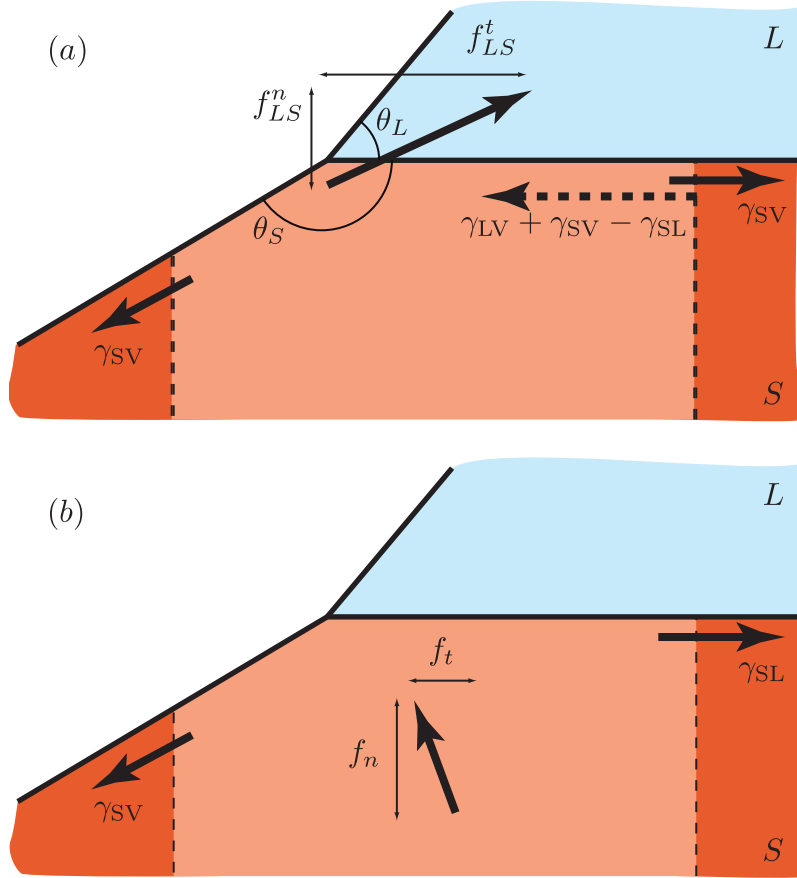


FIG. 3: (a) Distribution of capillary induced forces inside the solid. (b) Equivalent representation allowing a distinction between the forces at the contact line, and the forces that could be compensated by a Laplace pressure when the interfaces are curved.

where u_i is the displacement vector, and E the Young's modulus. This solid is assumed to be submitted to the long-range attractive potential. Then it can be rigorously shown (see the proof below) that the external solution must have an effective boundary condition:

$$\vec{\sigma}_s = (\Phi_{LS} + \Phi_{SS} - \Phi_{SL} - \Phi_{LL} + \Phi_{LL}^H(0) - \Phi_{SS}^H(0)) \vec{n}. \quad (39)$$

Note that this is the opposite of the surface force. When the liquid phase is expressed by a vapor phase, this expression reduces to:

$$\vec{\sigma}_s = (\Phi_{SS} - \Phi_{SS}^H(0)) \vec{n}. \quad (40)$$

The numerical solution presented in the main paper are based on numerical evaluations of the surface stress (39,40).

The surface stress σ_s acts as a generalized capillary pressure. At distances from the contact line much greater than the molecular cut-off a , the curvature is small so that [using (37,40)] the capillary pressure on the dry part reduces to the standard Laplace pressure:

$$\vec{\sigma}_s = \gamma_{SV} \kappa \vec{n}. \quad (41)$$

On the other hand, the Laplace pressure on the wet part is

$$\vec{\sigma}_s = \gamma_{SL} \kappa \vec{n}. \quad (42)$$

Proof – The incompressibility condition reads $\partial_l u_l = 0$ or in vector notations:

$$\vec{\nabla} \cdot \vec{u} = 0.$$

We first assume that the constitutive equation holds in the interfacial zone where the elastomer is subjected to two long range volumetric interactions which derive from the potentials Φ_{LS} and Φ_{SS} that are associated to liquid-solid and solid-solid interactions respectively. The equilibrium condition reads:

$$-\vec{\nabla}(p_r + \Phi_{LS} + \Phi_{SS}) + E\vec{\nabla}^2\vec{u} = 0. \quad (43)$$

Φ_{LS} and Φ_{SS} result from an integration over the volume of the liquid and the solid. They vary over a scale of few molecular sizes along the normal to the solid interface and attain their bulk values at distances away from the interface. Therefore, Φ_{LS} and Φ_{SS} are only important in a thin boundary layer across the interface. We pose $\tilde{p} = p_r + \Phi_{LS} + \Phi_{SS}$ and $\tilde{\sigma}_{ij} = \sigma_{ij} - (\Phi_{LS} + \Phi_{SS})\delta_{ij}$. Then the equation reduces to:

$$-\vec{\nabla}\tilde{p} + E\vec{\nabla}^2\vec{u} = 0 \quad (44)$$

in the whole domain. This is the standard elasticity problem. However the effective boundary conditions are modified. More precisely, there is no excess quantity in the strain tensor, but the effective stress applied to the bulk is modified. As the real stress σ_{ij} is continuous, the pseudo-stress $\tilde{\sigma}_{ij}$ is not.

Vectorial force transmission model

We can alternatively propose a model of elastomeric surface that allows for a fully vectorial transmission of the liquid-on-solid force. The polymeric chains are attracted in the direction of the liquid and thus transfer the total force exerted by the liquid on the solid. There is no force gradient building up in the surface layer. Integrating the equilibrium equation from the surface to the bulk, we see that the effective stress perfectly balances the excess forces due to the long range interactions:

$$\begin{aligned} \vec{\sigma}_s = & \vec{t} \cdot \vec{\nabla} \left[\int_0^\infty \Phi_{LS} dh \vec{t} \right] \\ & + (\Phi_{LS} + \Phi_{SS} - \Phi_{SL} - \Phi_{LL} + \Phi_{LL}^H(0) - \Phi_{SS}^H(0)) \vec{n} \end{aligned} \quad (45)$$

This is the expression for σ_s used in the numerical solution of the vectorial force transmission model.

Again, looking at a distance from the contact line large compared to a and using

$$\vec{t} \cdot \vec{\nabla} \vec{t} = -\kappa \vec{n},$$

we get in the dry region a surface stress:

$$\vec{\sigma}_s = (\gamma_{SV} + \gamma) \kappa \vec{n} \quad (46)$$

Beside, the Laplace pressure in the dry part is still

$$\vec{\sigma}_s = \gamma_{SV} \kappa \vec{n}. \quad (47)$$

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