Soft wetting and the Shuttleworth effect, at the crossroads between thermodynamics and mechanics

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Abstract – Extremely compliant elastic materials, such as thin membranes or soft gels, can be deformed when wetted by a liquid drop. It is commonly assumed that the solid capillarity in “soft wetting” can be treated in the same manner as liquid surface tension. However, the physical chemistry of a solid interface is itself affected by any distortion with respect to the elastic reference state. This gives rise to phenomena that have no counterpart in liquids: the mechanical surface stress is different from the excess free energy in surface. Here we point out some striking consequences of this “Shuttleworth effect” in the context of wetting on deformable substrates, such as the appearance of elastic singularities and unconventional capillary forces. We provide a synthesis between different viewpoints on soft wetting (microscopic and macroscopic, mechanics and thermodynamics), and point out key open issues in the field.

In this Perspective article we analyse the Shuttleworth effect from different viewpoints, and unify thermodynamic and mechanical approaches (Fig. 1). We discuss the consequences of $\Upsilon \neq \gamma$ in soft condensed matter are largely unknown [46].

Fig. 1: Perspectives on the Shuttleworth effect. Thermodynamics involves free energy minimisation, while the language of mechanics is expressed in terms of force balance. Macroscopically, these respectively involve the free energy per unit area $\gamma$, and the interfacial force per unit length $\Upsilon$. Microscopic equivalents, describing the molecular scale are given by Density Functional Theory (DFT) and Molecular Dynamics (MD, image from [47]).

|       | thermodynamics | mechanics |
|-------|----------------|-----------|
| macro | $F = \gamma \, A$ | $\Upsilon = \gamma + \frac{d\gamma}{dc}$ |
| micro | DFT            | MD        |
conditions under which it influences wetting of deformable media (Fig. 2), and point to open questions.

**Hierarchy of length scales.** – Before discussing the Shuttleworth effect, it is important to assess the various regimes of elastico-capillarity. Such a classification can be made in terms of the relevant length scales \[ \frac{B}{\gamma} \]. Consider slender elastic bodies, whose thickness \( h \) is much smaller than both its radius of curvature \( \kappa^{-1} \) and its length \( L \), such as that shown in Fig. 3a, depicting a drop of size \( R \) supported by a membrane of thickness \( h \ll R \). Viewed at the scale of the drop, the thin membrane deforms sharply near the edge of the drop, forming well-defined contact angles. Zooming in near the contact line, however, the membrane angle varies gradually. Owing to the membrane’s finite bending rigidity \( B \sim Eh^3 \), where \( E \) is the Young’s modulus, the bending occurs over a typical distance \( \kappa^{-1} \sim \left( B/\gamma \right)^{1/2} \). This length is referred to as the bending-elasto-capillary length. It provides, for instance, the characteristic size of the loop shown in Fig. 2. However, the subsequent zoom in Fig. 3a reveals a second length: the stretching-elasto-capillary length \( \gamma/E \). This is the scale over which the elastic solid deforms into a “wetting ridge” in the direct vicinity of the contact line.

The importance of the stretching-elasto-capillary length \( \gamma/E \) becomes apparent when the elastic body is not slender. This is further highlighted for drops on very soft elastomers or gels (Fig. 3b). Here one needs to introduce the “wetting ridge” in the direct vicinity of the contact line. Consider slender elastic bodies, whose thickness \( h \) is much smaller than both its radius of curvature \( \kappa^{-1} \) and its length \( L \), such as that shown in Fig. 3a, depicting a drop of size \( R \) supported by a membrane of thickness \( h \ll R \). Viewed at the scale of the drop, the thin membrane deforms sharply near the edge of the drop, forming well-defined contact angles. Zooming in near the contact line, however, the membrane angle varies gradually. Owing to the membrane’s finite bending rigidity \( B \sim Eh^3 \), where \( E \) is the Young’s modulus, the bending occurs over a typical distance \( \kappa^{-1} \sim \left( B/\gamma \right)^{1/2} \). This length is referred to as the bending-elasto-capillary length. It provides, for instance, the characteristic size of the loop shown in Fig. 2. However, the subsequent zoom in Fig. 3a reveals a second length: the stretching-elasto-capillary length \( \gamma/E \). This is the scale over which the elastic solid deforms into a “wetting ridge” in the direct vicinity of the contact line.

The importance of the stretching-elasto-capillary length \( \gamma/E \) becomes apparent when the elastic body is not slender. This is further highlighted for drops on very soft elastomers or gels (Fig. 3b). Here one needs to introduce the range of molecular interactions \( a \) as yet another length scale, setting the microscopic width of the interface. The three sequences of Fig. 3b show a double transition of the contact angles \[ \gamma \sim \omega \]. First, the microscopic contact angles change when \( \gamma/E \sim a \) [from panel (i) to (ii)], without affecting the apparent angle on the scale of the drop. The macroscopic angles only change when \( \gamma/E \sim R \) [from panel (ii) to (iii)]. These two transitions can be viewed as changes from Young’s law to Neumann’s law, respectively for the microscopic and macroscopic angles \[ \gamma_1 \sim \omega \sim \sigma \]. It should be noted, however, that there still is no microscopic derivation of Neumann’s law for elastic substrates that includes the Shuttleworth effect.

**The Shuttleworth equation.** – The surface energy \( \gamma \) is the excess free energy per unit area of an interface. An area \( A \) is therefore associated with an energy \( \gamma A \). Applying the virtual work principle to an increase in interfacial area \( \delta A \), one deduces the excess force per unit length \( T \). For a liquid interface, this leads to an increase \( \gamma \delta A \) of the surface free energy. Equating this change in energy to the mechanical work done by \( T \), one obtains the identity \( T = \gamma \). For liquids there is thus no need to distinguish between \( T \) and \( \gamma \) and one simply refers to surface tension. For elastic interfaces, however, the situation is fundamentally altered: expansion-induced strain changes the molecular structure of the interface. Hence, the interfacial excess free energy \( \gamma \) is not constant any longer, and we find a change in free energy

\[
\delta(\gamma A) = \left( \gamma + A \frac{d\gamma}{dA} \right) \delta A = \left( \gamma + \frac{d\gamma}{d\epsilon} \right) \delta A. \tag{1}
\]

where \( \epsilon \) is the strain parallel to the interface. Equating this to the work done by the surface stress, we find the Shuttleworth relation \[ \gamma \sim \omega \] (36,37):

\[
T = \gamma + \frac{d\gamma}{d\epsilon} \tag{2}
\]

In the context of soft matter, the derivative is understood as taken at constant chemical potential and temperature. The Shuttleworth effect gives rise to new phenomena that have no counterpart in liquids, whenever the surface free energy exhibits an explicit dependence on the strain.

**Thermodynamics: Measuring the Shuttleworth effect.** – The first illustration of the Shuttleworth effect is provided in a macroscopic thermodynamic framework. A suitable geometry to measure experimentally the strain derivative \( \gamma' = \frac{d\gamma}{d\epsilon} \) consists of a slender, elastic plate or rod partially immersed in a liquid reservoir (Fig. 4a). This setup forms an elastic realisation of the classical Wilhelmy plate (49), normally used to measure liquid surface tension. As mentioned, the slender theory is derived under the assumption of a hierarchy of lengthscales

\[
\max(\gamma/E, a) \ll h \ll \left( \frac{B}{\gamma} \right)^{1/2}. \tag{3}
\]

In this asymptotic limit, the spatial extent of the wetting ridge \( \gamma/E \) is confined to a small region near the contact line, and its effect on the global energy of the plate is negligible. Hence, we are in the same hierarchy of scales as for
and solid-vapor areas are represented by integrals over the

\[ \int A \] 

Here \( A \) is the liquid-vapor area, while the solid-liquid and solid-vapor areas are represented by integrals over the

\[ \int dA \] 

Data from [53].

Experimental measurement of the vertical displacement \( u \) along an elastic rod (\( \gamma/E \sim 1 \mu m, \text{radius} = 150 \mu m \)). The discontinuity of strain \( \epsilon = du/dz \) is clearly visible: the top part of the rod is stretched, the bottom part is compressed. Data from [53].

Intermediate zoom of the membrane in Fig. [3]: the conclusions thus equally apply to the membrane, even though we consider the plate in Fig. [4] under conditions where it remains straight [50].

The strain away from the contact line (distances \( \gg h \)) is homogeneous, though we need to distinguish the strain in dry part of the plate (\( \epsilon_+ \)) and the immersed part (\( \epsilon_- \)) – of Fig. [4]. The macroscopic free energy reads [51][52]:

\[ \mathcal{E} = \gamma_{LV} A_{LV} + \int_{\text{wet}} dA \left[ 2\gamma_{SL}(\epsilon_-) + \frac{1}{2} Eh\epsilon_-^2 \right] - F_{ext} z_{top} + \int_{\text{dry}} dA \left[ 2\gamma_{SV}(\epsilon_+) + \frac{1}{2} Eh\epsilon_+^2 \right]. \tag{4} \]

Here \( A_{LV} \) is the liquid-vapor area, while the solid-liquid and solid-vapor areas are represented by integrals over the

\[ \int dA \] 

“wet” and “dry” part of the surface – factors 2 reflect the two sides of the plate. Importantly, we allow for a dependence of the solid surface tensions on the strain \( \epsilon \); this will give rise to the Shuttleworth effect. Other terms represent the bulk elasticity, as well as the work done by the external force \( F_{ext} \). We neglect bulk swelling so that the reference state is well defined.

The elasto-capillary equilibrium of the wire is obtained by minimisation of the free energy [51][52]. In the Methods section, we briefly summarise the key steps of the derivation which can be performed for small strain, given the hierarchy of length scales. Variations of the contact line position and of \( z_{top} \), respectively, give two classical relations: Young’s law for the contact angle \( \theta \), and the force on the Wilhelmy plate, \( F_{ext} \sim \gamma_{SV} - \gamma_{SL} = \gamma_{LV} \cos \theta \). Variations with respect to \( \epsilon_+ \) and \( \epsilon_- \) involve the derivative of the surface energy \( \gamma' \) and lead to a new elasto-capillary coupling (see Methods):

\[ \epsilon_+ = \frac{2(\gamma_{SV} - \gamma_{SL})}{Eh} = \frac{2\gamma_{LV} \cos \theta}{Eh} \tag{5} \]

\[ \epsilon_- = \frac{2(\gamma_{TS} - \gamma_{SL})}{Eh}, \tag{6} \]

where the reference state is the plate surrounded by air.

Remarkably, the upper part of the plate probes the surface energies, while the immersed part probes the surface stresses. As a consequence, there is an elastic singularity in the form of a strain discontinuity when crossing the contact line region [51][52],

\[ \Delta \epsilon = \epsilon_+ - \epsilon_- = \frac{2(\gamma_{SV} - \gamma_{SL})}{Eh}, \tag{7} \]

which directly quantifies the Shuttleworth effect.

The discontinuity in strain has indeed been measured experimentally on a thin elastomeric rod. Figure [4] shows the vertical displacement field measured on a wire of polyvinylsiloxane partially immersed in ethanol [53]. With respect to the reference state — the rod surrounded by air — the upper part of the rod is stretched (\( \epsilon_+ > 0 \)), and the measured strain is in perfect agreement with the axisymmetric analogue of [4]. The lower part, however, is compressed (\( \epsilon_- < 0 \)). The discontinuity in strain can
thus be used to quantify the strength of the Shuttleworth effect, which in the experiment gives \[53\]
\[
\gamma_{SV}' - \gamma_{SL}' = 43 \pm 10 \text{ mN m}^{-1}
\]  
(8)
The magnitude of these terms is even larger than the relevant surface energies, \(\gamma_{LV} = 22.8 \pm 0.2 \text{ mN m}^{-1}\) and \(\gamma_{SL} - \gamma_{SV} = 16 \pm 4 \text{ mN m}^{-1}\). Hence, for this material, the influence of the Shuttleworth effect in elasto-capillarity cannot be considered a small correction.

**Macroscopic force balance near the contact line.** — We now turn to a mechanical view on the Shuttleworth effect. In the “slender body” description of the extensible rod, the strain discontinuity \([7]\) implies a perfectly localised line force of magnitude \(\gamma_{SL}' - \gamma_{SV}'\), near each of the two contact lines. However, this slender formulation does not reveal the mechanics on the scale of the thickness \(h\), let alone on the scales \(a\) and \(\gamma/E\). To gain insight in the force balance near the contact line, we now define a control volume of “mesoscopic” size \(w\) around the contact line. This control volume is indicated as the circle in Fig. 4 and further detailed in Fig. 5. Its size \(w\) is taken according to hierarchy of scales
\[
\max (\gamma/E, a) \ll w \ll h.
\]  
(9)
The first inequality ensures that the substrate remains essentially flat when viewed on the scale \(w\): this is important since in this limit the macroscopic contact angle \(\theta\) still obeys Young’s law.

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**Thermodynamics versus mechanics.** — We wish to emphasise that the “mechanical” result \([10]\) is fully consistent with the “thermodynamic” strain discontinuity \([7]\). The strain discontinuity that appears on the scale of the elastic rod \((\gg h, \text{Fig. 4})\) can be attributed to a tangential force \(F_\text{el}''\) generated near the contact line \((\ll h, \text{Fig. 5})\). Only when there is no Shuttleworth effect, one finds \(F_\text{el}'' = 0\) and a continuous strain across the contact line. The very same conclusion was recently drawn for a drop on a membrane \([54]\): the Shuttleworth-induced discontinuity of strain implies a jump in the membrane tension across the contact line, altering the contact angles on the scale of the drop (Fig. 3).

**Microscopic origin of the Shuttleworth effect.** — The experimental evidence of a significant Shuttleworth effect in a cross-linked polymer network (Fig. 4) comes as a surprise. Namely, one would have expected the structure of the surface at atomic scale to be close to that of an incompressible liquid, for which \(\Upsilon = \gamma\). By contrast, for hard crystalline materials, the microscopic physics of the Shuttleworth effect is more easily understood \([57]\), e.g. from a toy model consisting of a network of masses and springs. Due to redistribution of electronic charge in the vicinity of the surface, the effective properties of the springs (rest length, spring constant) are different in the surface from the bulk. Then, the interfacial zone naturally exhibits an excess elastic stress \(\Upsilon \neq \gamma\).

How can we understand the Shuttleworth effect for the liquid-like molecular structure of a cross-linked polymer network? In the continuum framework of Density Functional Theory (DFT), in the sharp interface approximation, it has been been possible to relate the Shuttleworth effect to a compressibility of the interfacial layer \([55]\). The corresponding physics is summarised in Fig. 4. Panel (a) shows a liquid phase (top) and an elastic phase (bottom) that are separated by a large vapour layer that can effectively be treated as a vacuum. The respective surface stresses are \(\Upsilon_{LV} = \gamma_{LV}\) and \(\Upsilon_{SV} = \gamma_{SV} + \gamma_{SV}'\). Panel
relations measured the total force that the liquid molecules plate, intended as a model for an AFM tip [56]. The simulations.
Figure 7a shows a simulation of a rigid Wilhelmy
confirmed quantitatively in Molecular Dynamics (MD) simu-
The importance of the work of adhesion has been con-
Eq. (12) provides two important predictions [55]:
description of the molecular structure at the interface,
While the DFT model represents a highly simplified
While joining the solid-liquid phases, this attraction leads
to an extra compressive stress in the interfacial zone in the
horizontal direction. However, this is not the case when the
interfacial layer is compressible: only a fractionνs/1−νs in terms of an interfacial Poisson ratio νs. Sum-
the vertical attraction leads to a compression of the interfacial
zone: the Shuttleworth effect arises whenever a fraction
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zone. The vertical attraction leads to a compression of the interfacial
layer is compressible: only a fraction
After joining the solid-liquid phases, this attraction leads
to an extra compressive stress in the interfacial zone in the
After joining the solid-liquid phases, this attraction leads
to an extra compressive stress in the interfacial zone in the
normal direction. For an incompressible layer this extra normal compression is equally transmitted in the tangen-
tial direction. However, this is not the case when the in-
compressible layer this extra normal compression is equally transmitted in the tangen-
tial direction. The vertical attraction leads to a compression of the interfacial

Hence, using (2) and (11), the microscopic model predicts

\[ \gamma'_{SL} - \gamma'_{SV} = (1 - \alpha)W. \]  
(12)

Fig. 6: Microscopic view of the Shuttleworth effect, based on
an approximate DFT model [55]. (a) A liquid and solid phase
are initially separated by a large vacuum. The surface stress
is computed as the excess force due to the missing molecular
interactions, exerted on the shaded region to the left of the
dashed line. (b) Joining the liquid and solid phases releases
(per unit area) the work of adhesion $W = \gamma_{LV} + \gamma_{SV} - \gamma_{SL}$. The vertical attraction leads to a compression of the interfacial
zone: the Shuttleworth effect arises whenever a fraction $\alpha < 1$
is transmitted horizontally.
While the DFT model represents a highly simplified
description of the molecular structure at the interface,
Eq. (12) provides two important predictions [55]:
- the departure from liquid-like behaviour relates to the
compressibility of the interfacial zone,
- the work of adhesion gives an upper bound on the
Shuttleworth effect ($\alpha = 0$):

\[ F_{cl}^d = \gamma'_{SL} - \gamma'_{SV} < W = \gamma_{LV}(1 + \cos \theta). \]  
(13)

The importance of the work of adhesion has been con-
firmed quantitatively in Molecular Dynamics (MD) simu-
lations. Figure 7a shows a simulation of a rigid Wilhelmy
place, intended as a model for an AFM tip [56]. The simu-
lations measured the total force that the liquid molecules
exert on the solid near the contact line: a large tangen-
tial force component was found, with a strength consistent
with the work of adhesion $\gamma_{LV}(1 + \cos \theta)$ [56], as predicted
in [57]. The often claimed $\gamma_{LV} \cos \theta$ is clearly not observed.
A similar MD simulation of a deformable Wilhelmy plate
showed a strain discontinuity, which using [7] gave a Shut-
tworth effect close to the upper bound set by the work
of adhesion [13] [51]. Finally, also the experimental value
for the elastomeric wire, quoted in [8], is very close to the
upper bound.
A final striking demonstration of the Shuttleworth ef-
fect is given in Fig. 7a. The picture represents a snapshot of a bubble on a weakly deformable wall, with the hierar-
chy of length scales $\gamma/E \ll a \ll R$. The contact angle is
close to $90^\circ$, which implies $\gamma_{SL} \approx \gamma_{SV}$. Despite this symmetry in surface energies, the elastic deformation is very
asymmetric [51]: the black arrows in Fig. 7a represent the
displacement field inside the solid, clearly showing a
strong tangential displacement towards the exterior of the
bubble. The breaking of symmetry is due to $\gamma_{SL} \neq \gamma_{SV}$,
even though $\gamma_{SL} \approx \gamma_{SV}$. The bias towards the liquid side
is perfectly in line with $F_{cl}^d = \gamma'_{SL} - \gamma'_{SV} \sim W$.

Fig. 7: The Shuttleworth effect in Molecular Dynamics. (a) A rigid probe partially immersed in a Lennard-Jones liquid. The simulations measure a large tangential force $\approx W$ on the solid
near the contact line. Image from [56]. (b) A gas bubble in a Lennard-Jones liquid on a deformable elastic solid, in a case
where $\gamma_{SL} = \gamma_{SV}$. The black arrows show the displacement
field inside the solid: despite the symmetry in surface energies,
the displacements are biased towards the liquid side due to
$\gamma_{SL} \neq \gamma_{SV}$. Data from [51]

Perspective. – Interfacial effects of soft solids pro-
vide an opening playground in soft condensed matter [46].
We have presented here some fundamental aspects of the
coupling between elasticity and capillarity – focusing on the so-called Shuttleworth effect that arises when surface free energy depends explicitly on the elastic strain. While we focussed on the prototypical “liquid drop on elastic solid”, the same issues and subtleties arise for adhesion of very soft solids [58, 62].

Figure 4 exemplifies a system (polyvinylsiloxane in contact with ethanol) that presents a strong strain-dependence $dγ/dϵ$. By contrast, no Shuttleworth effect was needed to accurately describe the wetting behaviour of water on polydimethylsiloxane [29,31]. A key open issue is therefore to understand the physicochemical conditions for the appearance of the Shuttleworth effect. Is it important that the liquid is a good rather than a bad solvent, as suggested by the interpretation in terms of surface compressibility? A more detailed understanding of the interface of a reticulated polymer with a liquid, and its consequences for elasto-capillary mechanics, is necessary. For example, we have ignored here the distance between crosslinks or entanglement points, which determines the scale at which entropy dominated elasticity and reference state are defined in the continuum. There is yet another length scale, associated with distinction between bulk swelling [63, 64] and interfacial effects associated with polymer free ends at the free surface of the sample. There is an urgent need for a systematic, quantitative characterisation of polymers and liquids, using a reliable set-up to measure the Shuttleworth effect. Can one design an alternative method to completely decouple from the Shuttleworth effect? A more detailed understanding of the interface of polymers and interfacial effects associated with polymer free ends would be needed to accurately describe the wetting behaviour of a liquid drop on a soft gel in the presence of the Shuttleworth effect. However, the detailed examples given in this paper relied on the stretching-elasto-capillary length $γ/E$ being relatively small. A systematic, fully consistent analysis still remains to be done for the more interesting cases of highly deformed interfaces. To give a striking example, even the selection of the contact angle for a droplet on a soft gel in the presence of the Shuttleworth effect is still an open problem on which contradictory statements can be found in the literature. Undoubtedly, soft elastic interfaces will continue to stretch our intuition for capillarity.

Methods. – The derivation of the strain discontinuity (7) involves some subtle kinematics: the contact line position $z_\text{cl}$ and the top of the plate $z_\text{top}$ are not independent of the strains $ε_\pm$. Here we briefly sketch the essential steps, which are properly developed in [51, 52]. First, we remind that the external force (per unit plate width) is $F_{\text{ext}} = 2(γ_{SV} - γ_{SL})$, which can be derived from a vertical displacement $δz_{\text{top}}$ at constant $ε_\pm$. The factor 2 is due to the two sides of the plate. Next, we consider variations $δε_\pm$ while keeping the contact line position fixed. The lengths of the dry/wet parts can be written as $L_\pm(1 + ε_\pm)$, where $L_\pm$ are lengths in the reference state. Expressing $z_{\text{top}} = z_\text{cl} + L_+(1 + ε_+)$, the energy per unit width (4) becomes

$$E = L_+(1 + ε_+) \left[ 2γ_{SV}(ε_+) + \frac{1}{2} E h_+^2 - F_{\text{ext}} \right]$$

$$+ L_-(1 + ε_-) \left[ 2γ_{SL}(ε_-) + \frac{1}{2} E h_-^2 \right].$$

(14)

where we omitted redundant constants. Minimisation now reduces to $dE/dε_\pm = 0$, which gives the equilibrium conditions (for $|ε_\pm| \ll 1$):

$$2(γ_{SV} + γ'_{SV}) + Eh_+ = F_{\text{ext}} = 2(γ_{SV} - γ_{SL})$$

$$2(γ_{SL} + γ'_{SL}) + Eh_- = 0.$$  

(15)

The combination $γ + γ'$ emerges in the same manner as in \[1\], where we derived the Shuttleworth equation for the surface stress. Combining the two equilibrium conditions yields the strain discontinuity (7). Equations (15) are obtained after subtracting the reference strain in air $ε_0 = - (γ_{SV} + γ'_{SV}) = - Υ_{SV}.$

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