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Statics and Dynamics of Soft Wetting

Bruno Andreotti¹ and Jacco H. Snoeijer²

¹Laboratoire de Physique de l'Ecole Normale Supérieure (LPENS), CNRS UMR 8023, Ecole Normale Supérieure, Université PSL, Sorbonne Université, and Université de Paris, 75005 Paris, France; email: andreotti@lps.ens.fr

²Physics of Fluids Group, Faculty of Science and Technology, and Mesa+ Institute, University of Twente, 7500AE Enschede, The Netherlands

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Abstract

The laws of wetting are well known for drops on rigid surfaces but change dramatically when the substrate is soft and deformable. The combination of wetting and the intricacies of soft polymeric interfaces have provided many rich examples of fluid–structure interactions, both in terms of phenomenology and from a fundamental perspective. In this review we discuss experimental and theoretical progress on the statics and dynamics of soft wetting. In this context we critically revisit the foundations of capillarity, such as the nature of solid surface tension, the microscopic mechanics near the contact line, and the dissipative mechanisms that lead to unexpected spreading dynamics.

Neumann's law:

vectorial balance of the three surface tensions at the contact line, traditionally used for liquid subphases

Wetting ridge:

localized elastic deformation of the substrate below the contact line

Viscoelastic braking:

slowing down of contact-line motion due to dissipation inside a viscoelastic substrate

Surface tension:

excess surface force per unit length; often called “surface stress,” but here we strictly reserve the term “stress” for the bulk force per area

1. INTRODUCTION: FROM RIGID TO SOFT WETTING

A liquid drop sitting on an ordinary solid does not induce any significant deformation of the surface. The angle made at the contact line is selected by surface energies according to Young's law. By contrast, a liquid drop floating on another liquid will strongly deform the interface, with contact angles selected by Neumann's law. Soft compliant substrates, typically reticulated polymer networks, are in between these two extreme cases: They do deform under the effect of capillary forces, but in contrast to liquids, they exhibit an elastic resistance.

The systematic exploration of soft wetting phenomena is fairly recent, spurred by improving technology to tune the properties of soft matter and by progress on elastocapillary phenomena in general—the latter received an extensive review by Bico et al. (2018). The mechanics of extremely soft materials originates from a competition between bulk elasticity and surface effects (Mora et al. 2010, Style et al. 2017), providing a new playground for material design. Applications are numerous and range from adhesives (Autumn et al. 2000, Boesel et al. 2010, Jagota & Hui 2011) to slippery surfaces (Lafuma & Qu  r   2011, Wong et al. 2011, Schellenberger et al. 2015, Solomon et al. 2016), highly stretchable synthetic materials (Grandgeorge et al. 2018), and the biomechanics of cells and soft tissues (Manning et al. 2010).

Here, we focus on the wetting of soft elastic substrates, for example by liquid drops. Apart from being of intrinsic interest, drops offer a unique way to study static and dynamic deformations of soft interfaces owing to their nanometrically sharp contact line forcing. Over the years, experiments such as those shown in **Figure 1** have progressively revealed the salient features of the wetting ridge below the contact line. This ridge dramatically alters the macroscopic spreading dynamics, as the moving ridge induces strong viscoelastic dissipation inside the substrate (Carr   et al. 1996, Long et al. 1996, Karpitschka et al. 2015, Zhao et al. 2018a). It leads to phenomena such as viscoelastic braking and dynamical depinning, which have no counterparts on rigid surfaces. Further complexities are encountered when the polymeric substrate swells by absorbing the wetting liquid (Cohen Stuart et al. 2006, Kajiya et al. 2011, Dupas et al. 2014, Boulogne et al. 2015) and when dangling chains are present at the surface.

These soft wetting phenomena are not captured by the same laws as rigid wetting (Andreotti & Snoeijer 2016, Style et al. 2017), and one is forced to critically revisit the foundations of capillarity, most notably, (a) the nature of surface tension of soft solids and the underlying microstructure of reticulated polymers, (b) the force balance near the contact line and the wetting boundary conditions, (c) moving contact lines and dissipation mechanisms inside the substrate, and (d) the resulting macroscopic motion of droplets. The purpose of this review is to address these fundamental aspects of wetting on soft surfaces by discussing recent experimental and theoretical progress.

2. BASIC CONCEPTS

2.1. Soft Polymeric Materials

For ordinary solid materials, externally applied stresses change the interatomic distances and thereby increase the internal energy. The resulting elasticity is of enthalpic origin and leads to elastic moduli of typically 10–100 GPa. Conversely, for reticulated polymer networks that are not in a glassy state, an applied strain puts chains in a less probable conformation, increasing the free energy only for entropic reasons. These networks can therefore undergo large reversible deformations at relatively small applied stress and present small elastic moduli, proportional to the thermal energy $k_B T$ and to the number of chains per unit volume [pieces of polymer between cross-links or between points of entanglement (Watanabe 1999)]. As such, the stiffness of the network can be varied over orders of magnitude via the density of cross-links or entanglements. A single polymer

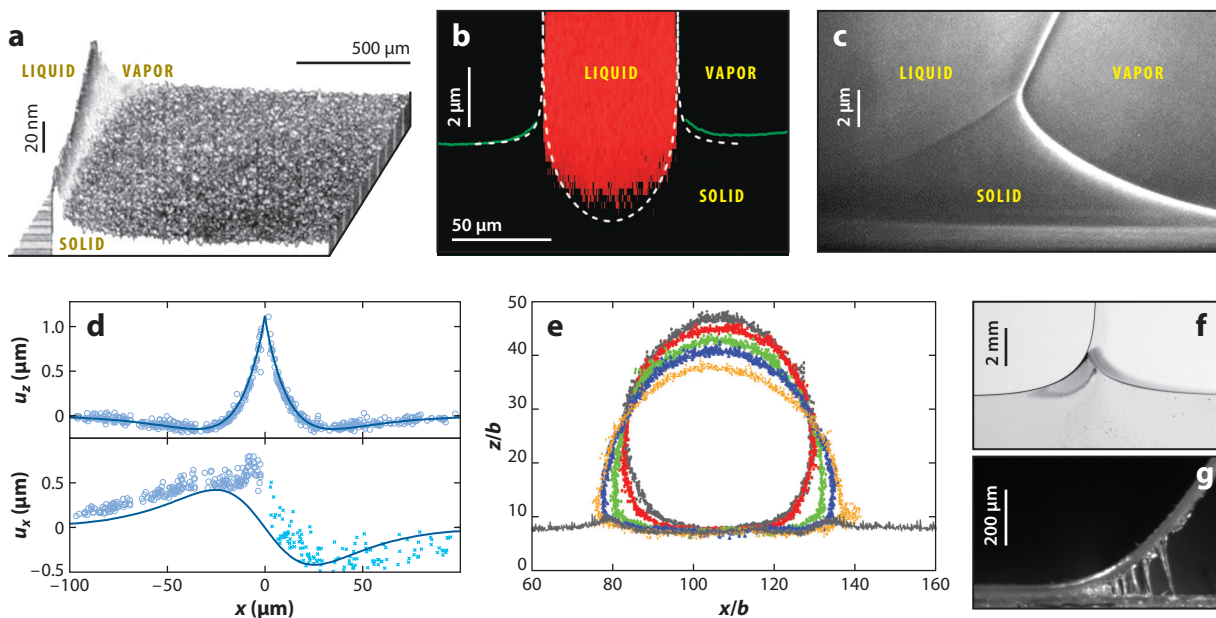


Figure 1

(a–d) Historical overview of wetting ridges observed on soft PDMS (polydimethylsiloxane) gels. The experimental methods used are (a) white-light interferometry, (b) laser scanning confocal microscopy, (c) X-ray, and (d) confocal microscopy. (e) Wetting ridges in molecular dynamics simulations of a polymer drop on a brush, for varying wettability. (f) Peeling of a tape from a PDMS gel (Perrin et al. 2019). (g) Peeling of a tape from a pressure-sensitive adhesive—note the fibril formation at the peeling front (Villey et al. 2015). Panels adapted with permission from (a) Carré et al. (1996), copyright 1996 Springer Nature; (b) Pericet-Camara et al. (2008), copyright 2008 American Chemical Society; (c) Park et al. (2014), under CC-BY 4.0 license; (d) Jerison et al. (2011), copyright 2011 American Physical Society; and (e) Leonforte & Mueller (2011), copyright 2011 AIP Publishing.

chain has a mechanical response that depends on its length N : In the simple Rouse model, its spring constant scales as $k_B T / N b^2$, where b is the monomer length, and its relaxation time τ scales as $\zeta b^2 N^2 / k_B T$, where ζ is the monomeric friction coefficient controlling the diffusion (Boese & Kremer 1990).

Before discussing their wetting behavior, we first provide a very brief description of the different types of polymeric substrates that can be encountered. For a complete view on the statistical physics involved, we refer the reader to textbooks by de Gennes (1979), Doi & Edwards (1988), Rubinstein (2003), and Binder & Kob (2011).

The adhesive properties of polymers strongly depend on their molecular architecture, as illustrated by peeling experiments. **Figure 1f** shows a reversible adhesive with a smooth viscoelastic ridge (Perrin et al. 2019), while **Figure 1g** involves a pressure-sensitive adhesive that leads to the formation of elongated fibrils (Villey et al. 2015). The latter polymer has strong adhesive bonds (Deplace et al. 2009) and ubiquitous dangling ends that need to be pulled from the matrix during debonding (**Figure 2a**), in contrast to the former (**Figure 2b**). The energy required to fracture the two interfaces, Γ , is proportional to the chain length and to the surface density of chains, Σ (**Figure 2e**) (Creton et al. 1992), and can therefore be orders of magnitude larger than the capillary adhesion energy due to van der Waals interactions (de Gennes 1989, Raphael & De Gennes 1992).

The network structure also determines the viscoelastic bulk rheology. Of particular importance for soft wetting are polymer gels (**Figure 2b**). These exhibit a fractal structure, with chain lengths spanning from the length of the prepolymer to the size of the sample. By consequence,

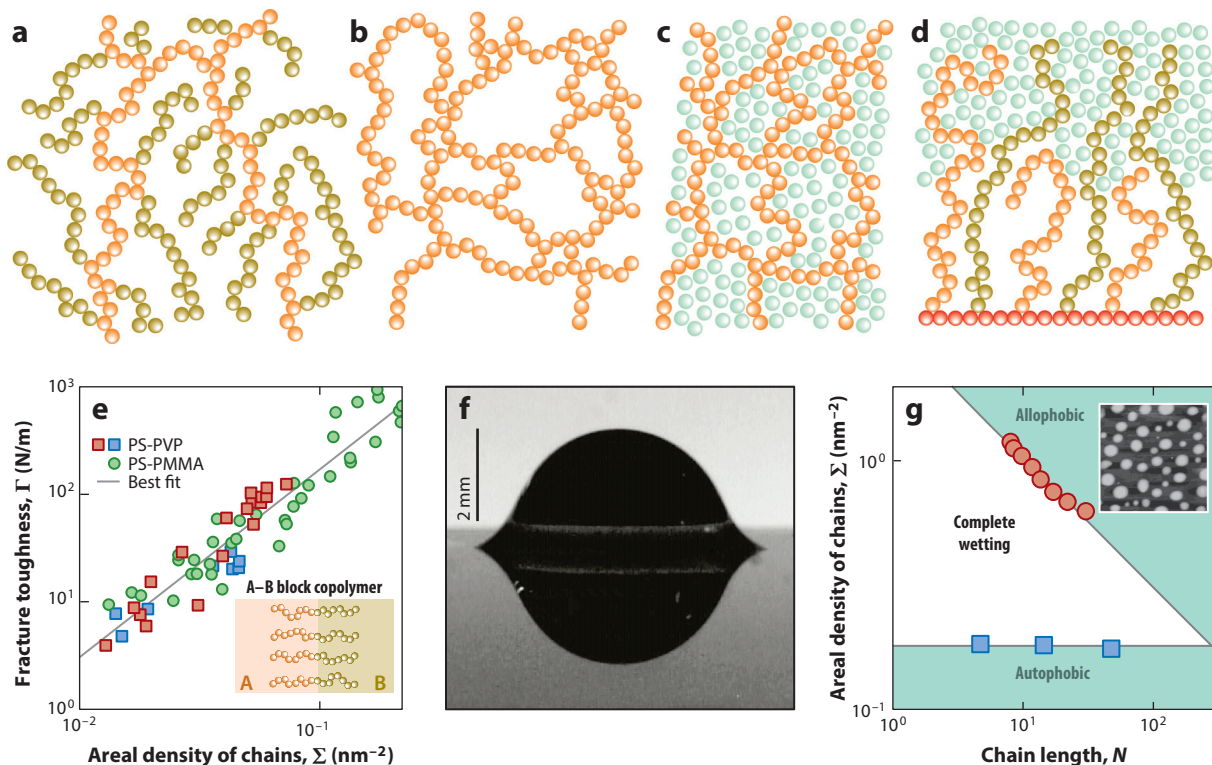


Figure 2

(Top row) Schematics of different architectures of polymeric soft solids: (a) a pressure-sensitive adhesive, characterized by many dangling chains (yellow) connected to a backbone network (red); (b) a polymeric gel, characterized by a multiscale network without dangling chains; (c) a swollen gel with a liquid phase; and (d) a polymer brush, here partly swollen. (e) The fracture toughness Γ of interfaces reinforced with a block copolymer plotted as a function of the effective areal density of chains, Σ . (f) A hydrogel sphere (with shear modulus $G = 61$ Pa) on a silicon wafer, totally wetted by water. (g) Wetting phase diagram of a melt of polystyrene of length N in contact with a brush of polystyrene chains end-attached to a substrate with a grafting density Σ . Solid lines are guides for the eyes through experimental points. Abbreviations: PS-PMMA, poly(styrene-*b*-methyl methacrylate); PS-PVP, poly(styrene-*b*-2-vinylpyridine). Panels adapted with permission from (e) Creton et al. (1992), copyright 1992 American Chemical Society; (f) Chakrabarti et al. (2018), copyright 2018 American Chemical Society; and (g) Maas et al. (2002), copyright 2002 American Chemical Society.

Storage $G'(\omega)$ and loss $G''(\omega)$ moduli:

the in-phase and out-of-phase stresses of a material under oscillatory strain, respectively; viscoelastic media exhibit both a reversible (storage) and a dissipative (loss) response

the relaxation spectrum spans orders of magnitude in frequency ω . This is reflected by the storage G' and loss G'' moduli, scaling as ω^n with an exponent $n \approx 0.5$ (Winter & Chambon 1986) close to that given by the Rouse model (Onogi et al. 1970). This viscoelastic response determines the substrate's dissipation in dynamical wetting experiments (de Gennes 1996, Long et al. 1996). When providing extra cross-links with respect to the gel point, the material exhibits a finite (static) shear modulus G at low frequency.

The substrate properties change dramatically when the network is swollen by a solvent (Figure 2c). A water drop placed on a hydrogel imbibes into the porous substrate, and one cannot always sharply define a contact line (Kajiya et al. 2011). A hydrogel sphere placed on a rigid surface, which is completely wetted by water, exhibits a finite contact angle (Figure 2f) (Chakrabarti et al. 2018). Polymer gels may even be swollen by a melt phase of the same polymer: Small fractions of un-cross-linked chains can alter the adhesive and dynamical properties (Jensen et al. 2015; Pham et al. 2017; Hourlier-Fargette et al. 2017, 2018). Swelling is also a key aspect

of brushes (**Figure 2d**), which are polymer chains tethered to a rigid substrate either covalently or by adsorption (Alexander 1977, de Gennes 1980, Milner et al. 1988). Brushes exhibit intricate wetting behaviors (Cohen Stuart et al. 2006, Leonforte & Mueller 2011, Mensink et al. 2019) and can even be autophobic with respect to their own melt (**Figure 2g**) (Maas et al. 2002).

Evidently, the physical chemistry of soft polymeric interfaces constitutes a vast area of research with numerous applications (Fleer et al. 1993). In the following, we primarily restrict ourselves to the statics and dynamics of wetting in the simplest case, where the polymer network is not swollen and where it does not present brush-like dangling chains. Part of the analysis will assume that pinning (contact angle hysteresis) is absent. This idealized situation can indeed be closely approached experimentally, e.g., by the frequently used PDMS (polydimethylsiloxane) gel substrates (**Figure 1**), although many other systems are touched on as well.

2.2. Capillarity: Liquid Versus Elastic Interfaces

From a macroscopic thermodynamic perspective, interfaces are characterized by a surface energy, γ . This represents the excess free energy per unit area of an interface (Rowlinson & Widom 1982, de Gennes et al. 2002). The order of magnitude of the surface energy is given by $\gamma \sim k_B T / a^2$, where the scale a is a typical microscopic length. Mechanically, this gives rise to a surface tension Υ_{ij} , which is the surface-analog of the stress tensor. It represents the excess force per unit length in the interface (Marchand et al. 2011); for a liquid, this tension is isotropic, $\Upsilon_{ij} = \gamma \delta_{ij}$. In what follows, the discussion concerns the two-dimensional case, for which we can stick to a scalar description of Υ . For an extensive review on mechanical implications of surface tension in soft solids, we refer the reader to Style et al. (2017).

Unlike liquids, for elastic interfaces the surface tension Υ and the surface energy γ are not equal, owing to the Shuttleworth effect (Shuttleworth 1950, Muller & Saul 2004). The origin of this difference is illustrated in **Figure 3**. **Figure 3a** shows that γ is the reversible energy associated with the creation of a solid–vapor interface by separating two solid blocks (i.e., not accounting for the fracture). This procedure is carried out at constant surface strain ϵ , although we remark that in general the surface energy can depend on strain, i.e., $\gamma(\epsilon)$. **Figure 3b** shows another way to create

Shuttleworth effect: the difference between surface tension Υ and surface energy γ , which arises whenever γ depends on the surface strain, ϵ

Surface strain: change in relative length of a surface element, measured with respect to the substrate's reference state

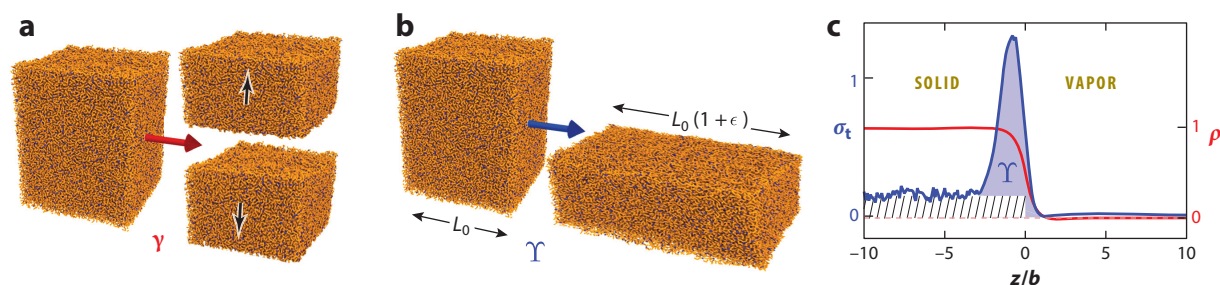


Figure 3

(a) The creation of two solid–vapor interfaces by dividing a solid block and bringing the interfaces far away from each other, while keeping the surface strain ϵ constant. The excess energy per unit surface area γ equals the reversible work done during the quasi-static separation. (b) The creation of a solid–vapor interfacial area by stretching the elastic solid. The change in interfacial energy equals the work done by the surface tension Υ and gives the Shuttleworth equation (Equation 1). (c) Molecular dynamics simulations of a reticulated polymer. (Red line) Profile of the monomer density ρ , indicating the location of the solid–vapor interface (the position is expressed in terms of monomer size b). (Blue line) Profile of the stress anisotropy, $\sigma_t = \sigma_{xx} - \sigma_{zz}$, across a stretched film. The effect of surface tension manifests itself as the peak of σ_t , located inside the liquid–vapor interface (the integral gives Υ). The hatched region is the bulk elastic stress. Data from Liang et al. (2018a,b).

Interfacial width:

capillary forces are not perfectly localized but spread out over a molecular region of size a between two bulk phases

Elastocapillary

length: length scale arising from the balance of surface tension and the shear modulus, setting the typical scale of wetting ridges

a solid–vapor interface, by stretching the interface length by $\delta L = L_0 \delta \epsilon$. The associated work per unit length, $\Upsilon \delta L$, involves surface tension Υ . Equating this work to the increase in surface energy, $\delta(L\gamma) = \delta [L_0(1 + \epsilon)\gamma]$, one obtains the Shuttleworth equation,

$$\Upsilon(\epsilon) = \frac{d}{d\epsilon} [(1 + \epsilon)\gamma(\epsilon)] = \gamma + (1 + \epsilon) \frac{d\gamma}{d\epsilon}. \quad 1.$$

The second term involves the derivative $\gamma' = d\gamma/d\epsilon$ and emerges due to the change in surface energy during stretching. This effect is not present for a simple liquid–vapor interface, for which we write $\Upsilon_{LV} = \gamma_{LV} = \gamma$.

It is of interest to examine the effect of surface tension at the nanoscale, inside the interfacial region (Weijs et al. 2013). The density profile across a soft interface exhibits a smooth transition over the molecular distance $a \sim 10^{-9}$ m (**Figure 3c**) (Liang et al. 2018a). Capillarity manifests itself as an anisotropy of stress components, in the direction tangential (σ_{xx}) and normal (σ_{zz}) to the interface. The stress anisotropy, $\sigma_t = \sigma_{zz} - \sigma_{xx}$, represents the excess tangential stress, or tension, localized in the interfacial zone. This effect is well known for liquids (Kirkwood & Buff 1949, Nijmeijer et al. 1990), but the concept equally applies for reticulated polymer networks. This is evidenced by the peak in σ_t (**Figure 3c**), whose integral gives the interfacial force per unit length: the macroscopic surface tension, Υ . Hence, the excess tangential stress inside the interface, $\sigma_t \sim \Upsilon/a$, can be estimated as $k_B T/a^3 \sim 10^7 - 10^8$ Pa. In crystalline and glassy solids, this stress is negligible with respect to elasticity, which is of enthalpic origin. By comparison, Υ/a is typically orders of magnitude larger than the entropic elastic modulus of a soft-polymer network, $G \sim k_B T/(Nb^3)$, owing to the large number of monomers N between cross-linkers. Since the monomer size b and the interfacial width a are both of molecular size, a crude estimate of the ratio $\gamma/G \sim Nb^3/a^2 \sim Na$ gives the so-called elastocapillary length.

We remark that **Figure 3c** was obtained from a molecular simulation of a simple cross-linked polymer network without solvent or dangling chains, for which one expects a liquid-like interfacial structure. The presence of dangling chains extends the interfacial region to the typical chain length, which requires a specific description at a mesoscopic scale.

2.3. Scales of Elastic Wetting

In a continuum perspective, elastocapillary phenomena can be classified in terms of length scales (Schroll et al. 2013, Bico et al. 2018). The elastocapillary length γ/G separates the small scales dominated by capillarity from the large scales dominated by elasticity. By tuning the substrate stiffness, γ/G can be varied over orders of magnitude, from submolecular up to millimeter scales. This length must be compared to the nanometric interface width a and to the macroscopic parameters such as the drop size R and the thickness e of the elastic substrate.

Figure 4a, subpanel *i*, illustrates the rigid limit ($\gamma/G \ll a$). The excess stress γ/a inside the liquid–vapor interface is then negligible compared to G and cannot induce any deformation below the contact line. Subpanel *ii* corresponds to the intermediate case ($a \ll \gamma/G \ll R$). Surface tension dominates at small scales and a sharp wetting ridge is formed (Jerison et al. 2011, Limat 2012, Marchand et al. 2012b). On the scale of the drop, however, elasticity is still dominant and the liquid angle remains unaffected. Subpanel *iii* illustrates the limit $\gamma/G \gg R$ where elasticity plays no role and the drop takes the shape of a liquid lens (Style & Dufresne 2012, Lubbers et al. 2014). An extensive discussion of the boundary conditions at the contact line, in particular of the applicability of Young’s law and Neumann’s law, follows in Section 3.

Figure 4b shows a drop on a thin membrane, in the regime $\gamma/G \ll e \ll R$. The membrane is curved over the bending elastocapillary length, $\sqrt{B/\gamma}$, which is based on the bending rigidity,

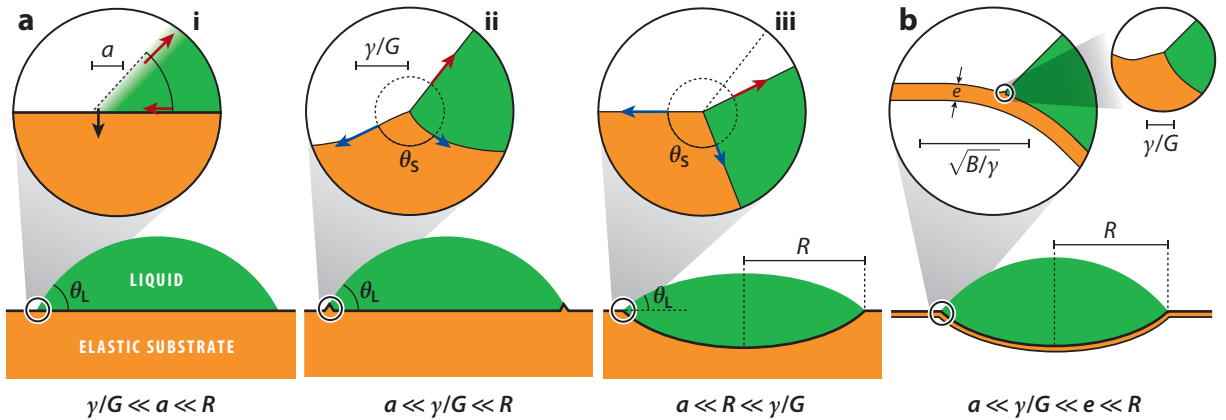


Figure 4

Scales of elastocapillarity. (a) Drops on thick elastic substrates of different stiffnesses, tracking the change of the liquid angle θ_L and of the solid angle θ_S (insets). The elastocapillary length γ/G must be compared to the width of the interface a and the drop size R . (i) The rigid limit, with contact angles governed by Young's law (see Marchand et al. 2011 for the force balance on the liquid wedge). (ii) Moderately soft substrates. The wetting ridge remains small compared to the drop size and θ_L still satisfies Young's law. However, θ_S is selected by Neumann's law, which expresses the balance of surface tensions applied to the circular system. (iii) The soft limit, where elasticity can be neglected on all scales. The drop resembles a liquid floating lens. (b) Typical case for a thin membrane of thickness e . The capillary-induced bending remains smooth at the bending length scale, $\sqrt{B/\gamma}$.

$B \sim Ge^3$. In that case, the wetting ridge is a negligible feature, and one enters the realm of elastocapillary bending and stretching of fibers and sheets (Roman & Bico 2010, Duprat et al. 2012, Schroll et al. 2013, Schulman & Dalnoki-Veress 2015, Bico et al. 2018, Davidovitch & Vella 2018).

3. THE WETTING BOUNDARY CONDITIONS

From a macroscopic perspective, interfaces can be treated as perfectly sharp. The effects of wetting then arise as boundary conditions at the contact line. Here we discuss the energetic derivation of these boundary conditions on elastic surfaces, in the absence of swelling by the wetting liquid. This is complemented by equivalent mechanical interpretations, which allow one to clarify the rigid-to-soft transition, and by a discussion of recent experiments.

3.1. Rigid Substrate Limit: Young's Law

To derive the classical Young's law on a rigid solid, one starts from the free energy $\mathcal{F}$ of the liquid–vapor interface of profile $b(x)$ whose contact line location is defined by the position $x = r$,

$$\mathcal{F} = \int_{-\infty}^r [\gamma(1 + b'^2)^{1/2} + \gamma_{SL}] dx + \int_r^{\infty} \gamma_{SV} dx. \quad 2.$$

The solid–liquid and solid–vapor surface energies respectively are γ_{SL} and γ_{SV} . Minimizing $\mathcal{F}$ with respect to variations of the interface profile $\delta b(x)$ and the contact line position δr yields, after integrating by parts,

$$\delta \mathcal{F} = \left[\frac{\gamma}{[1 + b'(x)^2]^{1/2}} + \gamma_{SL} - \gamma_{SV} \right]_{x=r} \delta r - \int_{-\infty}^r \gamma \kappa(x) \delta b(x) dx, \quad 3.$$

where $\kappa = b''/(1 + b'^2)^{3/2}$ is the interface curvature (Snoeijer & Andreotti 2008). The equilibrium condition $\delta\mathcal{F} = 0$ provides both the normal stress balance at the interface, $\gamma\kappa$ being the Laplace pressure, and the boundary condition at the contact line $x = r$, namely Young's law,

$$\gamma \cos \theta_L = \gamma_{SV} - \gamma_{SL}. \quad 4.$$

Any additional macroscopic force such as gravity or electrostatics acts as an additional normal stress along the interface but does not affect the boundary condition; the boundary condition can be interpreted as a horizontal force balance on an infinitesimal wedge of liquid near the contact line (Figure 4a, subpanel *i* inset).

3.2. First Boundary Condition: Neumann's Law for Contact Angles

When the substrate is soft enough to be deformed by the liquid, the elastic energy stored inside the substrate must be included in $\mathcal{F}$. Figure 5a defines the curvilinear coordinate s along the interface, the tangential vector $\mathbf{t}$, and the normal vector $\mathbf{n}$. The stress balance at the interface reads

$$\bar{\bar{\boldsymbol{\sigma}}} \cdot \mathbf{n} - \bar{\bar{\mathbf{T}}} \cdot \mathbf{n} = \frac{\partial}{\partial s} (\Upsilon_S \mathbf{t}), \quad 5.$$

where $\bar{\bar{\boldsymbol{\sigma}}} \cdot \mathbf{n} \equiv \boldsymbol{\sigma}$ is the elastic traction and $\bar{\bar{\mathbf{T}}} \cdot \mathbf{n}$ is the liquid traction; these are due to the stress tensors in the substrate $\bar{\bar{\boldsymbol{\sigma}}}$ and in the liquid $\bar{\bar{\mathbf{T}}}$, respectively. The surface tension of the solid is denoted Υ_S .

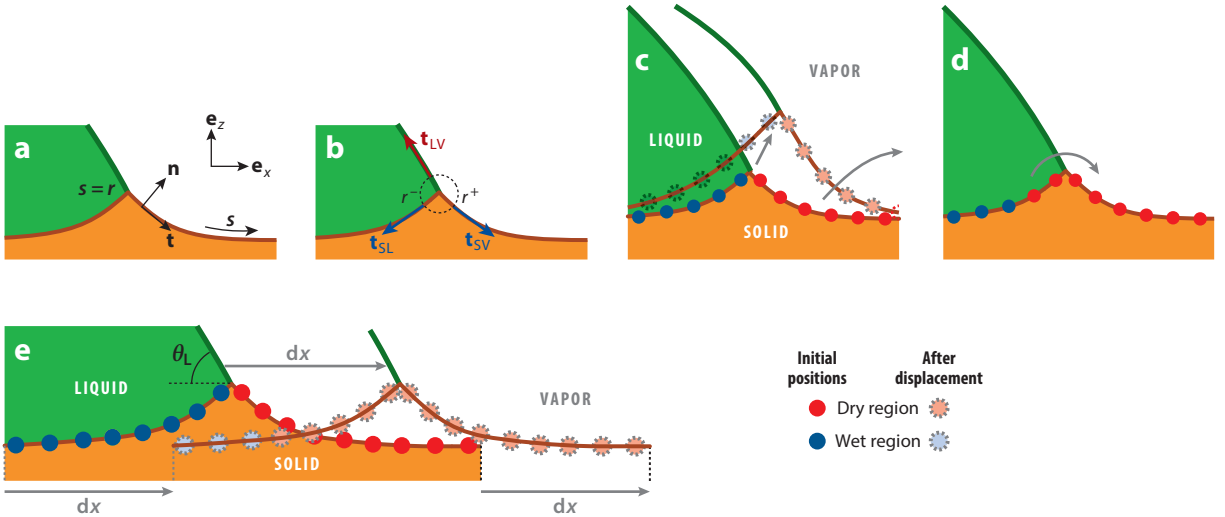


Figure 5

(a) Definition of the unit vectors $\mathbf{e}_x$ and $\mathbf{e}_z$ and the curvilinear coordinate s , running along the solid interface. The contact line is located at $s = r$ so that r_+ and r_- are respectively the limits on both sides. (b) Tangent unit vectors $\mathbf{t}^+ = \mathbf{t}_{SV}$ and $\mathbf{t}^- = \mathbf{t}_{SL}$ at both sides of the contact line. (c-d) Wetting at equilibrium requires the energy to be minimal with respect to all possible types of contact line displacement (Snoeijer et al. 2018). (c) Horizontal and vertical displacement in the lab frame. Equilibrium gives the first boundary condition (Equation 7), i.e., Neumann's law. (d) Relative motion of the substrate, while the contact line remains fixed in the lab frame. The colored points indicate the exchange of material points from the dry (red) to the wetted region (blue). Equilibrium gives the second boundary condition (Equation 9) on the chemical potential. (e) The liquid contact angle θ_L can be inferred from a global displacement dx of the contact line. The region close to the contact line remains unchanged, and the changes in (elastic and capillary) energy occur at the edge of the indicated contour.

The boundary condition at the contact line is again obtained from a variation of the contact line position. **Figure 5c** shows that this involves both horizontal and vertical displacements (Snoeijer et al. 2018). By consequence, there is now a vectorial boundary condition,

$$\gamma \mathbf{t}_{LV} + [\Upsilon_S \mathbf{t}]_{r^-}^{r^+} = \int_{r^-}^{r^+} ds \boldsymbol{\sigma}. \quad 6.$$

This can be interpreted as the integral of Equation 5 over an infinitesimal zone across the contact line (**Figure 5b**), using the perfectly localized liquid traction, $\overline{\mathbf{T}} \cdot \mathbf{n} = \gamma \mathbf{t}_{LV} \delta(s - r)$ (Limat 2012). When the substrate is forced into a wedge, the elastic stress is only logarithmically singular (Lubbers et al. 2014), also at large elastic deformation (Singh & Pipkin 1965). Given the weak stress singularity, the integral over $\boldsymbol{\sigma}$ vanishes and Equation 6 becomes

$$\gamma \mathbf{t}_{LV} + \Upsilon_{SV} \mathbf{t}_{SV} + \Upsilon_{SL} \mathbf{t}_{SL} = 0. \quad 7.$$

Figure 5b illustrates this result, known as Neumann's law, as clearly observed for the wetting of soft gels (**Figure 1**). This law is normally used for liquid phases (de Gennes et al. 2002) but also applies to sufficiently soft elastic substrates (Limat 2012, Marchand et al. 2012b, Style & Dufresne 2012, Style et al. 2013a). The weak elastic singularity of the (Cauchy) stress persists when prestretching the substrate, which therefore remains, contrary to recent suggestions (Masurel et al. 2018), integrable and does not contribute to Neumann's law (Snoeijer et al. 2018).

After these macroscopic considerations, it is instructive to consider the crossover from rigid to soft substrates from Equation 6. Microscopically, the liquid traction is not sharply localized in space but is spread out over the nanometric width a (**Figure 3c**). In the stiff limit, $\gamma/G \ll a$, the substrate remains flat and one recovers Young's law (Equation 4). In that case, Equation 6 can be used to compute the integral of elastic traction—integrating not over an infinitesimal zone, but over the contact line width, a . In the vertical direction this gives $\gamma \sin \theta_L$ to balance the upward pull of the droplet. This approach was already taken on by Rusanov (1975), Shanahan (1987), and White (2003) when computing the shape of the ridge for small deformation. Conversely, in the soft limit $\gamma/G \gg a$ where the solid forms a sharp wedge, the integrated elastic force per unit length T_{el} can be estimated as

$$T_{el} \sim \int_{r-a}^{r+a} \sigma ds \sim Ga \log\left(\frac{\gamma}{Ga}\right). \quad 8.$$

Molecular dynamics simulations indeed confirmed such an elastic correction to Neumann's law (Liang et al. 2018b). **Figure 6** reports T_{el} for both a droplet and a rigid particle, in contact with a cross-linked polymer network for a range of stiffnesses, here fitted with Equation 8. For experiments on soft wetting, γ/G is typically above the micron scale, for which the elastic correction to Neumann's law is less than one percent.

3.3. Second Boundary Condition: Surface Strain Discontinuity at the Contact Line

In contrast to the rigid limit, the substrate's elasticity allows for relative motion while the contact line remains stationary in the lab frame (**Figure 5d**). This motion involves an exchange of surface material across the contact line, indicating that the contact line is not pinned. This implies a second

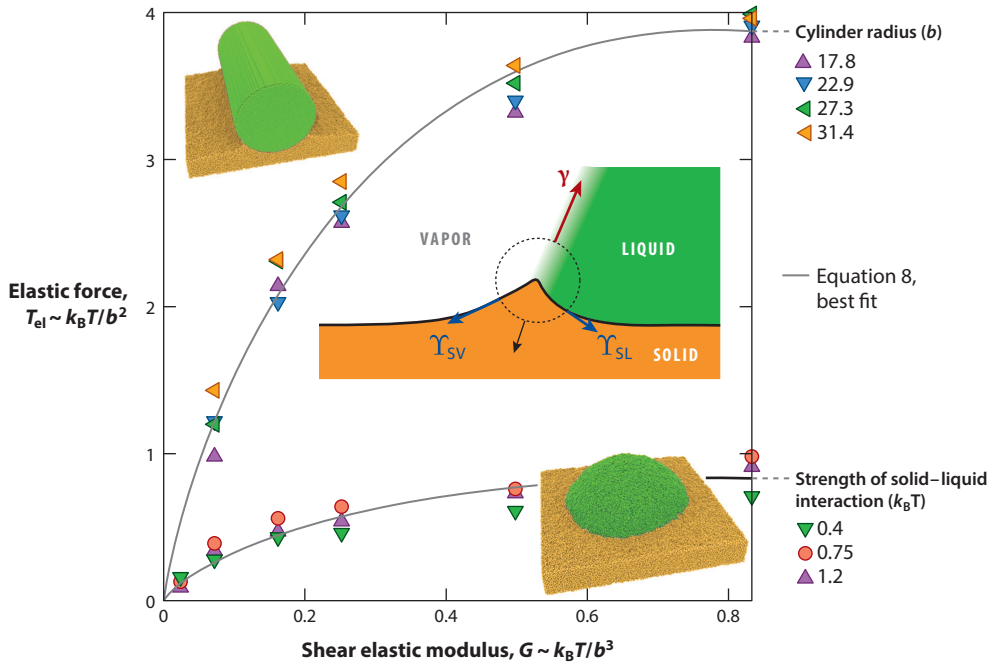


Figure 6

Rigid-to-soft transition on the microscopic scale. (*Inset*) The force balance on a control volume surrounding the contact line. The generalized Neumann's law involves the three surface tensions, γ , γ_{SV} , and γ_{SL} , and a residual elastic force per unit length, T_{el} . (*Main graph*) Elastic force T_{el} as a function of the shear elastic modulus G , measured in molecular dynamics simulations by Liang et al. (2018b). Data are represented in units of the thermal energy $k_B T$ and the monomer size b . Data are fitted by Equation 8, showing that T_{el} vanishes in the continuum limit $aG/\gamma \rightarrow 0$, where a is the molecular size and G is the shear modulus. Data from Liang et al. (2018b).

boundary condition (Snoeijer et al. 2018),

$$\mu_{SV} = \mu_{SL}, \quad \text{with} \quad \mu = (1 + \epsilon)^2 \gamma'(\epsilon) + f_{el} + \int_r^{r^\pm} (\boldsymbol{\sigma} \cdot \mathbf{t}) ds. \quad 9.$$

Here, f_{el} is the (surface) density of elastic energy that is associated to changing the material point below the contact line; it was shown to vanish in linear elasticity (Snoeijer et al. 2018) but possibly contributes at large strain. The integral over $\boldsymbol{\sigma}$ again vanishes in the soft limit ($\gamma/G \gg a$). The condition $\mu_{SV} = \mu_{SL}$ can be viewed as the equality of chemical potential that governs the exchange of material across the contact line. This is in direct analogy with equality of chemical potential across a liquid–vapor interface that regulates the discontinuity in density. Here, Equation 9 serves as a boundary condition for the surface strain ϵ , which in general can be discontinuous across the contact line.

In the rigid limit $\gamma/G \ll a$, any relative motion already implies a contact line displacement. The conditions of Equations 6 and 9 are then obtained from the variation of the same degree of freedom. In this rigid limit, the equality $\mu_{SV} = \mu_{SL}$ predicts the existence of a horizontal elastic force per unit length at the contact line, $\int (\boldsymbol{\sigma} \cdot \mathbf{t}) ds = \mathbf{e}_x \cdot \int \boldsymbol{\sigma} ds = \gamma'_{SV} - \gamma'_{SL}$; the same follows from Equation 6 in combination with Young's law (Weijs et al. 2013). This tangential force has indeed been observed in molecular simulations (Seveno et al. 2013).

THE MEMBRANE LIMIT

The membrane limit appears when the layer thickness e and γ/G are negligible with respect to the other scales (**Figure 4b**). The substrate is locally flat, and the elastic energy can then be integrated across the thickness. For small strains, the membrane limit gives an elastocapillary membrane energy per unit area, $\gamma(\epsilon) + Y\epsilon^2/2$, where Y is the membrane stiffness. Hence, Equation 9 simplifies to $\mu = \gamma' + Y\epsilon$, and the condition $\mu_{\text{SL}} = \mu_{\text{SV}}$ implies a strain discontinuity, $\epsilon_{\text{SV}} - \epsilon_{\text{SL}} = (\gamma'_{\text{SL}} - \gamma'_{\text{SV}})/Y$ (Weijs et al. 2013, Neukirch et al. 2014, Andreotti & Snoeijer 2016). This discontinuity is indeed observed in experiments and molecular simulations using an elastic Wilhelmy plate (**Figure 7a**). The liquid contact angle with respect to the membrane follows from Young's law based on the total membrane energy, and this gives $\gamma \cos \theta_{\text{L}} = \gamma_{\text{SV}} - \gamma_{\text{SL}} + Y(\epsilon_{\text{SV}}^2 - \epsilon_{\text{SL}}^2)/2$. Hence, the Shuttleworth effect implies an ϵ^2 -correction of the liquid angle (Weijs et al. 2013, Neukirch et al. 2014). The equations for the contact angle and the strain discontinuity provide the boundary conditions for the locally flat membrane (**Figure 3b**). The actual contact angle in the lab frame is obtained by solving the global membrane problem (Schroll et al. 2013, Davidovitch & Vella 2018).

3.4. Selection of the Liquid Contact Angle

The most prominent feature of wetting is the liquid angle θ_{L} with respect to the reference solid surface. It is not selected locally by the Neumann condition but emerges from the global, macroscopic elastocapillary problem of Equation 5 subjected to the two boundary conditions of Equations 6 and 9. The scenario for thick elastic layers is summarized in **Figure 4a**. For a drop of size R , the liquid angle undergoes a transition from Young's to Neumann's law—but this time on macroscopic scales, crossing over at $\gamma/G \sim R$.

Of particular experimental relevance is the regime $\gamma/G \ll R$, where the wetting ridge appears as a small feature on an otherwise flat substrate. In that case, θ_{L} can be derived from a global displacement of the contact line, as indicated in **Figure 5e**, comparing the surface and elastic energies at the edges of the contour, far away from the contact line. At the contact line, this global displacement involves a superposition of the motions in **Figure 5c,d**. Whether there is a correction to Young's law for θ_{L} depends on the elastic energy stored on both sides of the contact line, far from it. For free-standing membranes, the Shuttleworth effect can lead to a strain discontinuity, which gives an elastic correction to Young's law of order ϵ^2 (see the sidebar titled The Membrane Limit). For membranes glued to a rigid support, however, any jump in strain near the contact line will be screened by the finite membrane thickness. Hence, the strains far away from the contact line are equal, and θ_{L} follows from Young's law.

3.5. Is There a Shuttleworth Effect for Polymeric Solids?

The Shuttleworth effect is well established in crystalline materials (Muller & Saul 2004). For nonglassy polymer networks, however, it is less obvious that a change in the surface strain ϵ would alter the molecular structure of the interface, which is usually thought of as being close to that of an incompressible liquid. Hence, it is not clear a priori that soft polymeric substrates exhibit a strong dependence of surface energy γ on the applied strain ϵ . Although different numerical and theoretical works have been devoted to this question (Weijs et al. 2013, Liang et al. 2018b, Masurel et al. 2018), it is important in this review to focus on experimental results.

Experimental evidence for a strong Shuttleworth effect in soft wetting is provided in **Figure 7a**, which shows the elastic displacement inside a thin elastomeric Wilhelmy plate, partially immersed (Marchand et al. 2012a). The observed discontinuity of strain across the contact line implies

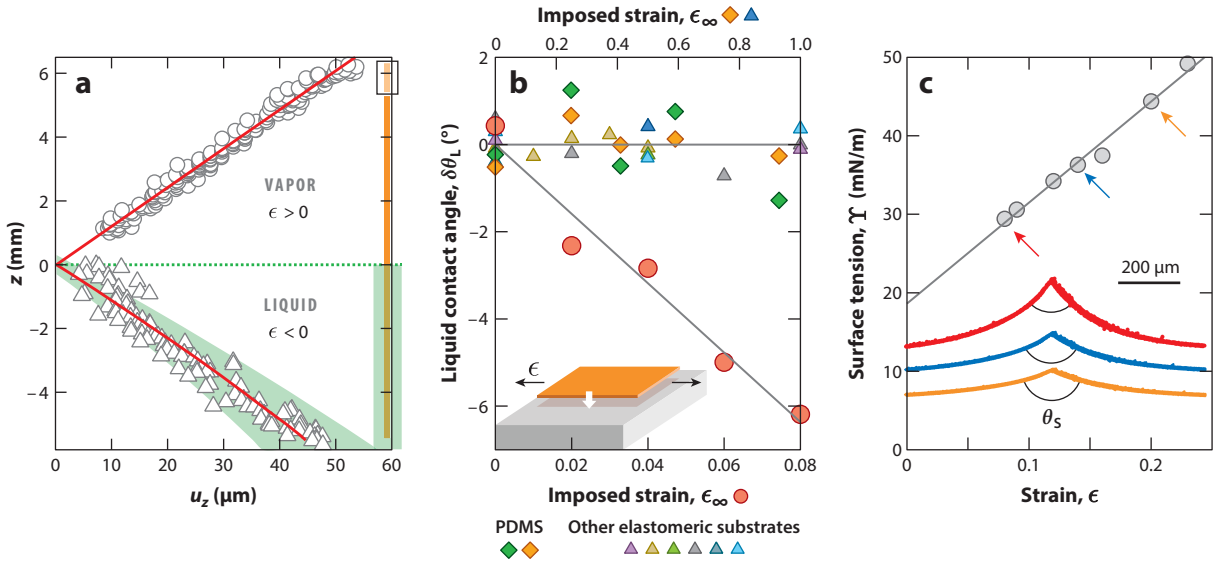


Figure 7

Soft wetting experiments investigating the Shuttleworth effect. (a) The elastic Wilhelmy plate, where an extensible rod (of elastocapillary length $\gamma/G \sim 1 \mu\text{m}$) is partially immersed in a liquid. The data represent the vertical displacements along the rod, showing a discontinuity of strain, $\epsilon = du_z/dz$, across the contact line. Data from Marchand et al. (2012a). (b) Change of the liquid contact angle $\delta\theta_L$ versus imposed strain ϵ_∞ . (Red circles; lower axis) θ_L on a stiff glassy polymer exhibits a strong dependence on strain. (Other symbols; upper axis) θ_L for drops on various soft elastomers exhibits no dependence on strain. Triangles correspond to various elastomeric substrates used in experiments by Schulman et al. (2018), and diamonds correspond to soft PDMS (polydimethylsiloxane) (advancing and receding angles) used in experiments by Snoeijer et al. (2018). (c) Dependence of solid surface tension Υ on strain ϵ on PDMS, as inferred from the change in the solid angle θ_s of the wetting ridge (inset). Data from Xu et al. (2017).

$\gamma'_{SV} - \gamma'_{SL} = 43 \pm 10 \text{ mN/m}$, which is comparable to the relevant surface energies. A strong contact angle hysteresis was present in these experiments.

A recent series of experiments has systematically investigated the contact angles obtained upon externally stretching the substrate (Schulman et al. 2018). **Figure 7b** reports changes in the liquid contact angle $\delta\theta_L$ as a function of the imposed strain ϵ_∞ (with negligible contact angle hysteresis). These experiments were carried out in the regime where θ_L follows Young's law, so that they directly investigate the strain dependence of $\gamma_{SV} - \gamma_{SL}$. For stiff glassy polymers, $\gamma/G \ll a$, there is a clear change of contact angle, and hence a strong Shuttleworth effect, observed for four different liquid–glass combinations (only one data set is shown). The other data correspond to drops on a broad variety of soft elastomers with $\gamma/G \gg a$. These exhibit no variation of θ_L , even for strains up to $\epsilon_\infty = 1$. This implies that $\gamma_{SV} - \gamma_{SL}$ does not depend on strain ϵ . This robust outcome, found for seven different liquid–elastomer combinations (Schulman et al. 2018, Snoeijer et al. 2018), can be interpreted as evidence either that there is no measurable Shuttleworth effect or that there is a fundamental reason why γ_{SV} and γ_{SL} share the same dependence on ϵ . The second boundary condition (Equation 9), which needs to be satisfied in the absence of contact line pinning, indeed requires $\gamma'_{SV} = \gamma'_{SL}$ in the regime of small deformations (Snoeijer et al. 2018). However, rigorous results for large deformations are currently lacking, and it is unknown why many polymeric gels would obey this property.

In another series of experiments on PDMS, the solid angle θ_s was measured as a function of the imposed strain ϵ_∞ (Xu et al. 2017). **Figure 7c** shows that θ_s becomes shallower with strain—even though θ_L remains constant. The lack of dependence of θ_s on drop size and substrate thickness

consolidates the interpretation of these experiments using Neumann's law (Equation 7) without any residual elastic tension, as mentioned in the discussion of Equation 8 above. The change of θ_s then implies a strong dependence of Υ on ϵ . This method also allows for a direct measurement of the tensorial nature of surface tension when applying anisotropic stretching (Xu et al. 2018).

A definite interpretation of these experiments will require the development of a fully nonlinear modeling approach and a consistent treatment of the Shuttleworth effect, including the second boundary condition (Equation 9). This needs to be complemented with a calibration of the material properties in the regime of large elastic deformations to account for the influence of stretching on the ridge shape.

4. DYNAMICAL ELASTOCAPILLARITY

4.1. Viscoelastic Braking

Pioneering studies by Shanahan & Carré (1995), Carré et al. (1996), and Long et al. (1996) revealed a dramatic slowing down of contact line motion when the substrate becomes exceedingly soft. The motion of the wetting ridge induces a time-dependent deformation of the substrate, leading to strong viscoelastic dissipation that opposes rapid motion. Typical velocities of millimeter-sized drops sliding under the influence of gravity can be as low as 10–100 nm/s (Karpitschka et al. 2016a) due to strong viscoelastic braking. A direct illustration of this effect occurs when drops move more rapidly over thin layers, as these induce less dissipation inside the substrate (Zhao et al. 2018a).

Figure 8a shows experimental data for a variety of dynamical wetting experiments. When the contact line moves with a velocity U , the contact angle changes from the equilibrium contact angle according to $\delta\theta_L \sim U^n$, with an exponent n close to 0.5. Linear viscoelasticity precisely predicts this behavior and relates the exponent n to that of the loss modulus, $G''/G \sim (\omega\tau)^n$ (Long et al. 1996). The contact line speed U excites the viscoelastic solid at a frequency $\omega \sim U/\ell$, where ℓ is the width of the ridge; at small velocities, ℓ is on the order of γ/G . A mechanical analysis on the scale of the wetting ridge shows that the contact line motion induces a rotation of the wetting ridge (Karpitschka et al. 2015), shown in the inset of **Figure 8a**. The associated dissipation in the solid follows as

$$\frac{1}{2} \int dx dy \left(\bar{\bar{\sigma}} : \bar{\bar{\gamma}} \right) \sim \ell^2 G'' \omega \sim \ell^2 G \left(\frac{U\tau}{\ell} \right)^n \frac{U}{\ell} \quad 10.$$

and is balanced with the injected power $\sim \gamma U \delta\theta_L$ due to capillary forces. When moving at a velocity U , the contact angle therefore changes from the equilibrium contact angle according to

$$\delta\theta_L \sim \left(\frac{U\tau}{\gamma/G} \right)^n. \quad 11.$$

This is in excellent agreement with experiments, both for advancing and receding motion (**Figure 8a**). Importantly, the dissipation is only integrable for $n \leq 1$ (Long et al. 1996). For $n = 1$, the standard moving contact line singularity for viscous liquids is recovered (Bonn et al. 2009, Snoeijer & Andreotti 2013). For an analogy between viscoelastic braking and dynamical adhesion, see the sidebar titled Dynamical Adhesion.

The argument can be made rigorous by deriving the shape of the wetting ridge from Equation 5, where the elastic traction σ is computed using a Green's function formalism (Johnson 1985). This Green's function approach has been extensively used to compute static wetting ridges (Jerison et al. 2011, Limat 2012, Marchand et al. 2012b, Style & Dufresne 2012, Bostwick et al. 2014, Lubbers et al. 2014) but can be extended to the dynamical case to account for the frequency

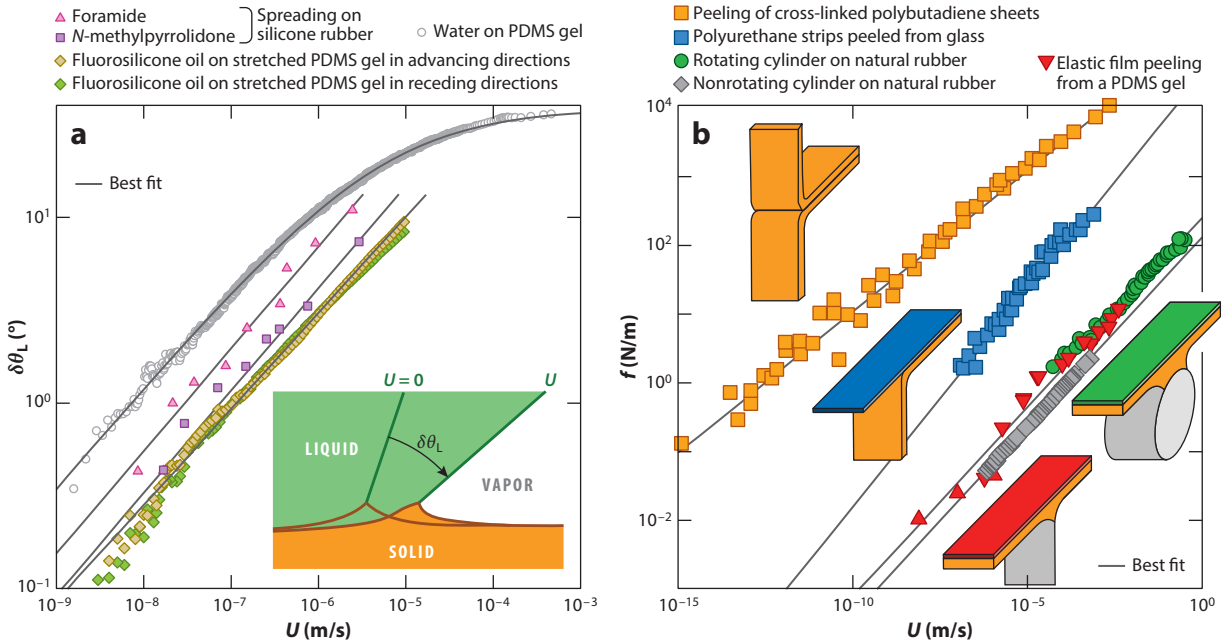


Figure 8

Energy dissipation inside a moving wetting ridge. (a) Variation of the liquid angle $\delta\theta_L$ as a function of the contact line velocity U in different liquid–solid wetting experiments: formamide (red triangles) and *N*-methylpyrrolidone (blue squares) spreading on silicone rubber (Shanahan & Carré 1995), water on a PDMS (polydimethylsiloxane) gel (white circles) (Karpitschka et al. 2015), and fluorosilicone oil on a stretched PDMS gel in the advancing (orange diamonds) and receding (green diamonds) directions (Snoeijer et al. 2018). The solid gray lines show the best fits by power laws, and the upper gray curve includes phenomenologically the saturation at large velocity. (b) Relation between the dissipative force f and velocity U in solid–solid adhesion experiments: peeling of cross-linked sheets of polybutadiene (orange squares) (Gent 1996); polyurethane strips peeled from glass (blue squares) (Maugis & Barquins 1978); a cylinder moving on natural rubber, allowed to rotate (green circles) (Charmet et al. 1995) or not (gray diamonds) (Robbe-Valloire & Barquins 1998); peeling of an elastic film from a PDMS gel (red triangles) (Perrin et al. 2019). The solids lines show best fits by power laws. Data provided courtesy of the cited authors and replotted here.

dependence of the storage and loss moduli. For small velocities, this linear response framework gives the same result (Equation 11) for the ridge rotation (Karpitschka et al. 2015). Hence, the liquid angle passively follows the ridge rotation and maintains a Neumann balance even in the dynamical case.

At larger velocities, however, the size of the wetting ridge decreases. This is because the effective stiffness increases at frequencies beyond τ^{-1} , giving rise to a dynamical elastocapillary length, $\ell \sim \gamma/|G' + iG''|$. The volume over which dissipation occurs is then diminished, leading to a saturation of Equation 11 at large velocities; $\delta\theta_L$ approaches a constant value (Figure 8a) at large velocities. The saturation is indeed observed above the viscoelastic velocity $\gamma/(G\tau)$ (Karpitschka et al. 2015), providing further direct evidence that the wetting dynamics is governed by substrate viscoelasticity.

4.2. Stick-Slip Dynamics

When a droplet is forced to move at high velocity, a remarkable stick-slip dynamics is observed (Pu & Severtson 2008; Pu et al. 2010; Kajiya et al. 2013, 2014; Karpitschka et al. 2015; Park et al.

DYNAMICAL ADHESION

Soft adhesives are polymeric layers that are used to bind rigid structures together; they must stick on most surfaces by simple contact under a low normal stress (Creton 2003). Their adherence strength is strongly dependent on the peeling velocity. Most pressure-sensitive adhesives exhibit a large nonlinear viscoelastic dissipation during debonding due to the formation of fibrils from cavitation (Teisseire et al. 2007, Villey et al. 2015) (**Figure 1g**), as observed using probe tack tests (Nase et al. 2008, Vilmin et al. 2010, Chopin et al. 2018). Polymer chains across the fracture are stretched until they store an elastic energy comparable to the covalent binding energy (Lake & Thomas 1967). A bond-breaking event dissipates all this stored energy, so that the fracture energy Γ is proportional to the length N of the chain and to the cross-link density Σ . Cavitation and fibrillation typically take place when the elasto-adhesive length Γ/G is larger than the adhesive thickness e (Amouroux et al. 2001, Deplace et al. 2009, Creton & Ciccotti 2016).

Conversely, reversible adhesives can be peeled without bulk cavitation or plastic deformations and have been the subject of recent interest in biomimetics (Autumn et al. 2000, Ghatak et al. 2004, Boesel et al. 2010, Jagota & Hui 2011). During fracture, the front exhibits a wetting ridge similar to that observed in soft wetting (**Figure 1f**), and the resulting dynamics indeed bears a strong resemblance. **Figure 8b** reviews the relation between the dissipative force per unit length and the contact line velocity for different fracture tests and different reversible adhesives. They indeed all exhibit a power law dependence that originates from the viscoelastic rheology (Schapery 1975, Newby et al. 1995, de Gennes 1996).

A deep connection between soft wetting and (reversible) adhesion has indeed been established over the last years, primarily in the static regime. Upon increasing the elastocapillary length, now given by γ/G , one can continuously go from the classical JKR (Johnson, Kendall, and Roberts) theory in solid mechanics to the normal case of liquid wetting (Salez et al. 2013, Style et al. 2013c, Hui et al. 2015, Cao et al. 2014). Surface tension in fact provides the small-scale regularization of the crack singularity observed in JKR (Johnson et al. 1971, Johnson 1985, Liu et al. 2014, Karpitschka et al. 2016b) and in dynamical situations controls the size over which dissipation takes place (Perrin et al. 2019).

2017; van Gorcum et al. 2018). **Figure 9a** shows the liquid angle as a function of time from a dip coating experiment (Kajiya et al. 2013). Importantly, the stick-slip motion is not associated with permanent defects of the substrate (Bonn et al. 2009, Snoeijer & Andreotti 2013) but here results dynamically from the self-induced wetting ridge. In solid friction, stick-slip occurs as an instability due to a decrease of the friction force when the velocity increases (Baumberger & Caroli 2006). The instability disappears at large velocity due to structural aging (Rice & Ruina 1983). Velocity weakening is also the mechanism invoked to explain the stick-slip observed during the peeling of adhesive tapes (Cortet et al. 2007, De Zotti et al. 2019), and a similar shear weakening leads to the formation of shear bands in complex fluids (Divoux et al. 2016).

In the dynamical wetting of soft solids, the friction force results from the viscoelastic loss due to the ridge motion. A velocity weakening mechanism would therefore relate stick-slip dynamics to the rheology. Indeed, stick-slip appears when the frequency of excitation due to the contact line motion becomes comparable to the rheological cross-over frequency τ^{-1} at which the storage and loss moduli become comparable. For paraffin gels, stick-slip is observed in an intermediate range of velocities (Kajiya et al. 2013, 2014). At high and low speeds, one recovers a continuous spreading, as in solid friction, corresponding to a purely viscous and a purely elastic response, respectively. For PDMS gels, however, a single transition from continuous to stick-slip motion has been reported (Karpitschka et al. 2015, Park et al. 2017, van Gorcum et al. 2018). The lack of continuous motion at high speed can be traced back to the PDMS rheology at high frequency, for which G' and G'' remain comparable for all relevant frequencies beyond τ^{-1} .

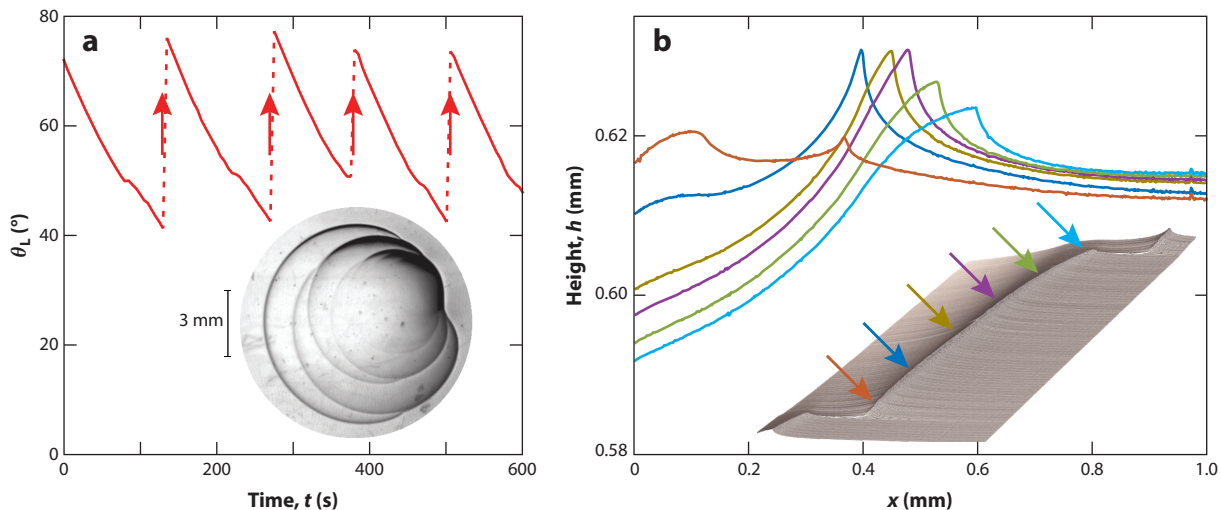


Figure 9

Stick-slip motion during forced spreading. (*a*, *inset*) Picture of a SBS–paraffin gel surface after the spreading of a drop whose stick-slip has left multiple circular patterns. Data from Kajiy et al. (2013). (*a*) Time evolution of the liquid angle θ_L in a dip-coating experiment, performed with the same substrate. The contact angle slowly decreases as it sticks to the substrate and suddenly increases during a slip event. Data from Kajiy et al. (2014). (*b*) Wetting ridge profile as a function of time during a stick-slip cycle, as the contact line is forced over a PDMS gel. Note that the tip angle of the wetting ridge increases when approaching the depinning event. (*Inset*) Space-time diagram. Abbreviations: PDMS, polydimethylsiloxane; SBS, poly(styrene-*b*-butadiene-*b*-styrene). Data from van Gorcum et al. (2018).

In the case of PDMS, however, friction does not decrease with velocity. Instead of a single wetting ridge that accelerates during slip events, direct visualizations have shown that the contact line depins from its own wetting ridge and surfs it (Park et al. 2017, van Gorcum et al. 2018). The depinning is governed by the Gibb's inequality that is normally used for sharp edges on rigid surfaces (van Gorcum et al. 2018) and is related to the increase of the solid angle of the ridge θ_s with contact line velocity (**Figure 9b**). This change of θ_s points either to time-dependent surface stresses or to dynamical effects beyond a simple static Neumann balance. Although for purely elastic media the wetting ridges decay after depinning, for some systems plastically deformed ridges remain after stick-slip events (Pu et al. 2010), confirming the generality of the depinning scenario.

4.3. Manipulating Droplet Motion

When exposed to a humid environment, the deformability of the substrate strongly affects the condensation of droplets (Sokuler et al. 2010) (**Figure 10a**). Similar features are observed on lubricant-infused surfaces (Kajiy et al. 2016), where instead of a gel layer the drop is in contact with a viscous lubricant that is maintained in a porous substrate (Schellenberger et al. 2015, Keiser et al. 2017). Both cross-linked polymer networks and lubricant surfaces can exhibit vanishing contact angle hysteresis, making them of particular interest for applications such as antifouling and food packaging (Wong et al. 2011, Solomon et al. 2016). In addition, the use of soft coatings can be highly effective for the suppression of splashing (Howland et al. 2016).

Once several drops are in close proximity, the substrate deformation leads to intricate interactions between adjacent drops (**Figure 10b**). This inverted Cheerios effect resembles the capillary interaction of solid particles at liquid interfaces (known as the Cheerios effect), except that the roles of liquid and solid are reversed. The interactions can be manipulated from attractive to

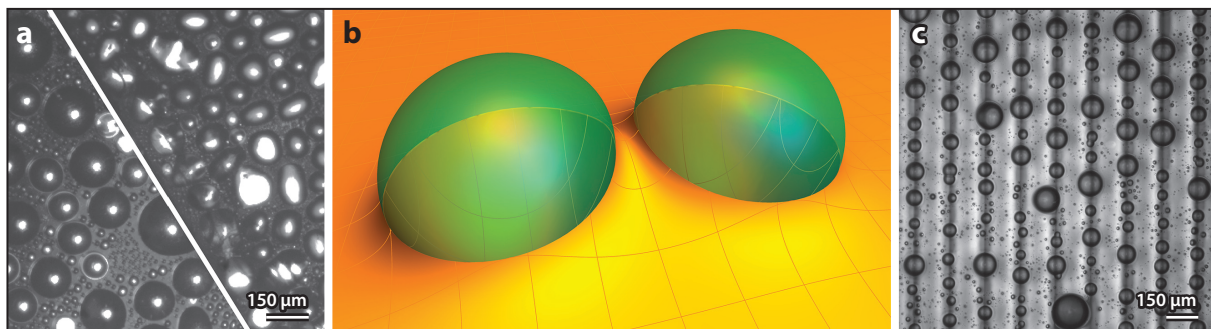


Figure 10

(*a*) Condensation on two PDMS (polydimethylsiloxane) surfaces cooled simultaneously under identical experimental conditions, the left one being five times softer than the right (Sokuler et al. 2010). (*b*) The inverted Cheerios effect: a three-dimensional calculation of the elastocapillary meniscus between two drops, leading to interactions (Karpitschka et al. 2016a). (*c*) Droplet durotaxis. Glycerol droplets, deposited on a flat surface with stiffness gradients, move toward the thickest regions of the substrate (Style et al. 2013b). Panel *a* adapted with permission from Sokuler et al. (2010), copyright 2010 American Chemical Society, and panel *b* adapted with permission from Style et al. (2013b).

repulsive by tuning the geometry of the substrate (Karpitschka et al. 2016a). A similar approach to manipulate droplet motion is achieved by imposing gradients in the mechanical properties of the substrate. Droplets exhibit durotaxis (Style et al. 2013b), resembling that of biological cells (Saez et al. 2007, Trichet et al. 2012), and tend to migrate toward soft regions (**Figure 10c**). Such a migration was observed for sliding drops, exhibiting a curved trajectory in the presence of gradients of dissipation, bending toward thicker regions (Zhao et al. 2018a).

4.4. Dynamical Wetting of Brushes and Swollen Gels

The continuum vision of the dynamical elastocapillary problem is perfectly adapted to the case of a totally reticulated polymer gel without dangling ends or residual melt content. In this final section, we report dynamical experiments on other systems, such as brushes and swollen gels. **Figure 2g** showed that brushes are highly effective in changing the wettability of the substrate (Maas et al. 2002) and can reduce contact angle hysteresis to within a few percent of a degree (Lhermerout et al. 2016). For dynamical wetting, the dissipation inside the substrate can again be the dominant factor, now due to the stretching and relaxation of the polymer chains. **Figure 11a** shows that the dissipative force (quantified by $\delta\theta_L$) in a PDMS brush is proportional to the contact line velocity, rescaled by the brush thickness ℓ and the relaxation time τ (Lhermerout et al. 2016). This opens up the promising perspective of using dynamic contact lines to probe the nanorheology of polymeric materials (Restagno et al. 2002, Garcia et al. 2016).

The swelling of a polymer gel by un-cross-linked chains may substantially modify the contact line dynamics: A capillary-induced phase separation can occur that extracts free chains from the cross-linked polymer network, which then accumulate at the surface and alter the wetting velocity (Hourlier-Fargette et al. 2017, 2018). Also without phase separation, the dynamics can be dominated by poroelastic effects, competing with viscoelasticity (Johnson 1982, Hong et al. 2008, Doi 2009). After removing the pulling force, the relaxation of wetting ridges may indeed be limited by poroelastic diffusion (Zhao et al. 2018b, Berman et al. 2019).

Figure 11b reports an experiment motivated by the food industry, where water drops spread on a soluble glassy polymer (Dupas et al. 2014). At low velocity, the change of the contact angle

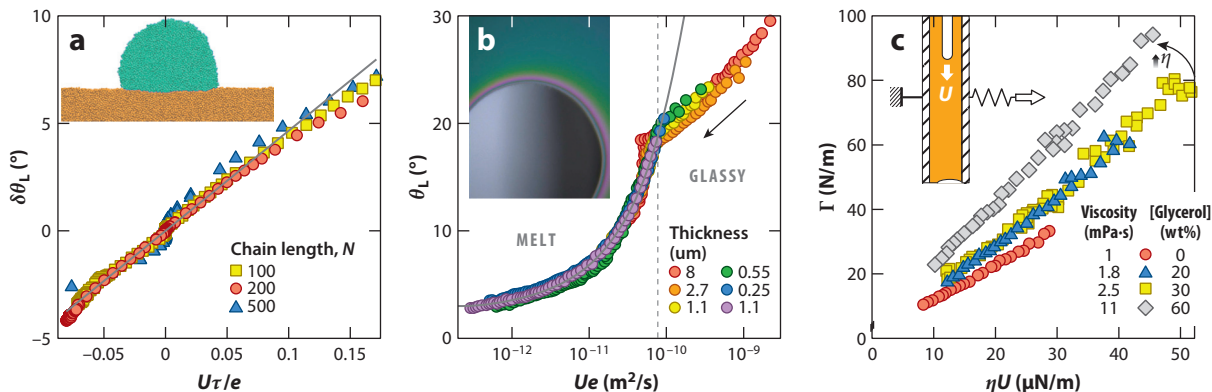


Figure 11

(a) Dissipation induced by a liquid contact line moving on PDMS (polydimethylsiloxane) pseudo-brushes, collapsing data for varying chain lengths. The graph shows the variation of the liquid contact angle $\delta\theta_L$ with the velocity U , rescaled by the thickness e of the brush and the Rouse relaxation time τ . (Inset) Droplet on a brush in a molecular dynamics simulation (Mensink et al. 2019). (b) Liquid angle θ_L as a function of the contact line velocity U for water droplets spreading on maltodextrin of different thicknesses e , from 100 nm to 8 μ m. (c) Fracture energy release rate Γ as a function of front velocity U for gels with the same gelatin concentration ($c = 5$ wt%) and various glycerol contents and therefore different viscosities η . Panels adapted with permission from (a) Lhermerout et al. (2016); (b) Dupas et al. (2014), copyright 2014 American Physical Society; and (c) Baumberger et al. (2006b), copyright 2006 Springer Nature.

is due to hydration of the substrate ahead of the contact line, resulting from the evaporation and subsequent condensation on the substrate. The water concentration depends on the product of the speed U and the layer thickness e and indeed leads to a collapse of data onto a master curve (Figure 11b). Above a critical velocity, the diffusion is too slow and the substrate remains in a glassy state. Note that the scaling with e is different in panels *a* and *b*, reflecting the different dissipative mechanisms at play. Further complexity is encountered when the droplet wetting the hydrogel contains solutes. A surfactant-laden drop can induce fracture of the gel (Bostwick & Daniels 2013, Grzelka et al. 2017), while the addition of particles can offer strategies for the deposition of particles to avoid the coffee stain effect, and even to tune the contact angle (Boulogne et al. 2015, 2016, 2017).

We have already mentioned the large fracture energy Γ for debonding pressure-sensitive adhesives, associated with pulling connecting chains from the matrix. Similar effects arise in the fracture of swollen physical gels, in which cross-linking is due to weak reversible bonds. Figure 11c shows that the crack velocity under a given load decreases when the solvent viscosity increases. In this case, the dissipation is dominated by viscoplastic chain pull-out (Raphael & De Gennes 1992), hence the dependence on chain/solvent friction (Baumberger et al. 2006a). This illustrates that further dissipative mechanisms should be anticipated when exploring soft wetting in a broader class of soft-polymer substrates.

FUTURE ISSUES

1. To turn soft wetting experiments into fully quantitative probes for (surface) rheology, researchers should include in future models large elastic deformations (van Brummelen et al. 2017, Masurel et al. 2018, Wu et al. 2018) and a fully consistent implementation of the Shuttleworth effect.

2. What are the physicochemical conditions determining the appearance of the Shuttleworth effect?
3. Under what conditions do soft substrates exhibit hysteresis? When can droplets induce viscoplastic deformations inside the substrate? Could these effects be included as a process zone at the wetting ridge, as in fracture mechanics?
4. What mechanisms could lead to the dynamical variation of θ_s (**Figure 9**), which initiates the depinning in the stick-slip regime?
5. Similar to lubricant-infused surfaces, the combination of surface topography and functionalization by polymers offers new routes toward surface engineering.
6. Contact line forces provide a unique mechanical probe of biological matter, while drops on gels offer a model system to study principles of mechanobiology (Schwarz & Safran 2013, Charras & Sahai 2014, Humphrey et al. 2014).

DISCLOSURE STATEMENT

The authors are not aware of any biases that might be perceived as affecting the objectivity of this review.

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