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Static and dynamic contact lines on a deformable coating of controled thickness.

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Introduction

Elastowetting phenomena, i.e. wetting on deformable substrates are presently attracting great interest [1]. It is well known that the normal component of liquid surface tension deforms the substrate and leads to the formation of a "ridge" at the contact line, which in turn affects drastically the static as well as dynamic wetting properties. While the static properties have received a lot of attention [1-2], comparatively much less is known for the dynamics [3-4], especially when the substrate is composed of a very thin layer deposited on a rigid substrate. This is in contrast with the large range of techniques that could benefit from this knowledge: development of anti-dew coating by interposition of a thin absorbing gel layer on a solid, control of wetting and adhesion on solids by gel layers, nutritive substrates for biophysics, etc. We have investigated this question, experimentally, by following the axisymmetric retraction of circular water drops, in which liquid is removed at constant flux rate, on thin silicone elastomer layers of controlled thickness bound to rigid glass slides. We have also developed models, for both the statics and dynamics, in terms of Green functions [2], that are in excellent agreement with the experimental data. This modeling effort has allowed us to evidence a new and simple law linking the dynamic contact angle to both the contact line velocity and the layer thickness [5], that holds for very thin layers of gels, before crossing over to a more classical behavior predicted by Long et al [3] in the 90's at larger thicknesses.

Experiments

We have prepared substrates by depositing thin layers of a silicone elastomer (PDMS Sylgard 527, Dow Corning, USA), of thickness varying from 8 µm to several millimeters, on rigid glass plates. We have checked with rheological measurements that this material is well described by the Chasset-Thirion model over the range of frequencies relevant to the experiments,, i.e. by a complex shear modulus given by: $G'(\omega)+i$ $G''(\omega) = \mu_0(1+i(\omega\tau)^m)$, with $\mu_0 = 1085 \pm 124$ Pa, m = 0.66 \pm 0.04 and $\tau = 15.4 \pm 0.4$ ms. Pure water drops of surface tension $\gamma \approx 72$ mN/m, were deposited on this coating, of radius 0.1 to 2 mm (see fig. 1-a). The static contact angle was measured by side view; it varies a bit upon drop radius (fig. 1-d), while remaining rather close to 90°, indicating that the substrate surface tension in the wetted and dry area are very close to have the same value called here γ_{s} . The details of surface deformation were explored with a Schlieren method that we have developed [5]. The typical structure that appears (Fig. 1-b) is composed of a "ridge" induced by the liquid surface tension that pulls the solid upward, and of a slight depression or "dimple" very near the contact line to guarantee mass conservation, the silicone gel being incompressible. Our Green function model (see details below) allowed us to recover exactly the profile (fig. 1-c), the sole fitting parameter being the surface tension of the coating, that we found to be $\gamma_8 \approx 40$ mN/m, in good agreement with previous experiments performed by other groups on the same kind of compound [1]. We then explored the dynamics by removing liquid from the drop at constant flux rate, following the evolution of both the contact angle and of the contact line velocity. The data are reproduced on fig. 1-e, and compared to a theoretical model that we have built by generalizing the dissipation calculations proposed by Long et al in 1996, to our situation (see below).



Figure 1: (a) side view of a drop deposited on the soft coating; (b) schematic of the details near contact line, (c) comparison of model and experimental profiles deduced from a Schlieren method; (d) static contact angles for "small" and "big" drops; (e) results obtained for the dynamical case, compared to modeling and, in the insert with the power law on the velocity deduced from eq. (3); (f) test with all data of our law (4), dotted line, while the law proposed in [3] is the continuous line.

Models.

To model the static distortions of the substrates, we modified a model that we had developed for infinite depth [2], solving Stokes equations obtained from incompressible elasticity equations with a zero-displacement imposed at the surface of the rigid basis. After using a potential function defined on the horizontal and vertical displacements as $u_x = -\partial \phi / \partial y$ and $u_y = \partial \phi / \partial x$, and working in the Fourier space, one gets the following expression for the surface vertical displacement induced by a straight contact line in its vicinity:

$$\zeta(x) = \frac{f_y}{2\pi\gamma_s} \int_{-\infty}^{+\infty} dk \cos kx \left[k^2 + \frac{\mu}{\gamma_s K(k)} \right]^{-1} + \frac{f_x}{2\pi\gamma_s} \int_{-\infty}^{+\infty} dk \sin kx \left(\frac{{h_0}^2 k^2}{\sinh 2h_0 k - 2h_0 k} \right) \left[k^2 + \frac{\mu}{\gamma_s K(k)} \right]^{-1}$$
(1)

in which $f_x = \gamma \cos \theta_e$ and $f_y = \gamma \sin \theta_e$, are the vertical and horizontal forces induced on soft layers by the liquid-vapor surface tension, and the quantity K(k) reads:

$$K(k) = \frac{1}{2k} \left[\frac{\sin 2h_0 k - 2h_0 k}{2h_0^2 k^2 + \cos 2h_0 k + 1} \right]$$
(2)

To mimic a circular drop of radius R, we identified the result near each contact line to the one of a rivulet of width 2R, having thus the same Laplace pressure enclosed inside. This leads to the curves of Fig. 1-c, in which the surface profiles are exactly recovered, provided that one uses the substrate surface tension extracted from the fit, i.e. $\gamma_S \approx 40 \text{ mN/m}$. To solve the question of the dynamic contact angle θ_{dyn} , we used the same method as above, assuming that the contact line, i.e. the point on which the forces $f_x = \gamma \cos \theta_{dyn}$ and $f_y = \gamma \sin \theta_{dyn}$ are applied is moving at a constant speed V, and replacing the elastic shear modulus with the Chasset-Thirion expression with $\omega = kV$. Having obtained the instantaneous displacement field, following Long et al.'s method [3], we calculated the energy dissipated in the coating and assumed that this energy was supplied by the work of the horizontal force $f_x = \gamma \cos \theta_{dyn}$. After calculations, this leads to the following law, valid at low velocity:

$$\frac{\left(\cos\theta_{dyn} - \cos\theta_{eq}\right)}{\left(\sin\theta_{dyn}\right)^{2}} \approx \left(\frac{\gamma}{\gamma_{s}}\right) \frac{\mu_{0}}{\gamma_{s}} |V\tau|^{m} \int_{0}^{\infty} \frac{k^{m+2}}{\left(k^{2} + \frac{\mu_{0}}{\gamma_{s}K(k)}\right)^{2}} dk$$
(3)

If one now considers the limit of very low thickness of the coating, this expression can be simplified and gives a simple analytical law:

$$G(\theta_{dyn}) = \frac{\left(\cos\theta_{dyn} - \cos\theta_{eq}\right)}{\left(\sin\theta_{dyn}\right)^2} \approx 2\left(\frac{\gamma}{\gamma_s}\right) \left(\frac{V\tau\mu_0}{\gamma_s}\right)^m \left(\frac{h_0\mu_0}{\gamma_s}\right)^{3(1-m)/4}$$
(4)

that we tested with success on fig. 1-e (dotted line at low values). At larger values of the argument, the result crosses over to another saturating value, announced long ago by Long et al [3]. To our knowledge, it is the first time that the regime (4) is identified experimentally and exactly calculated.

Conclusions

In summary, we investigated the statics and dynamics of wetting on soft, viscoelastic coatings deposited on a rigid surface. While the static contact angle does not seem to depend on the thickness of the soft layer, this parameter, on the contrary, has a key influence on the dynamic contact angle. The result is a bit counterintuitive as the contact line moves faster when the substrate becomes thinner, which is the reverse of what would be expected for a contact line moving on a liquid substrate. We also mention that our result does not coincide with the one of ref. 4, in this limit of very low thickness, for reasons that can be found in ref. [5]. More recently we focused on the case of finite deformations, with high substrate slope, in collaboration with R. Masurel and I. Ionescu (LSPM, Univ. Paris 13) [6]. We found that everything is to be reconsidered, as Neuman triangle becomes modified with an extra-force associated to the singular nature of the ridge. This, however does not modify the scaling laws that we have found for the dynamics.

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