Influence of spatial heterogeneities on spreading dynamics

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Abstract. Consider the motion of a two-dimensional droplet of a partially wetting fluid over topographical substrates. Assuming that spreading occurs in the Stokes regime, a singular perturbation procedure yields a set of integro-differential equations for the locations of the two droplet fronts. By analyzing these equations, we illustrate a number of intriguing features that are not present when the substrate is flat, such as the existence of multiple equilibria which allows for a hysteresis-like effect on the apparent contact angle. Additionally, we demonstrate a stick-slip behavior of the contact line as it moves along the local variations and the interesting possibility of a relatively brief recession of one of the contact lines.

1. Introduction

The influence of spatial heterogeneities on droplet spreading is arguably a much less theoretically studied subject compared to spreading over ideally flat substrates (e.g. [1, 2, 3, 4] and the references therein) even though they are known to impact the dynamics significantly regardless of their size. Specific spatial substrate variations may be desired in some manufacturing processes, but more often than not small-scale spatial defects are unavoidable and manifest themselves as ‘noise’ in the system. The corresponding substrates are commonly called ‘rough’ and have been the subject of the majority of experimental studies on the influence of spatial heterogeneities on spreading dynamics (e.g. Schwartz & Tejada [5] and Semal et al. [6]).

The few theoretical works on droplets over topographical substrates have been devoted primarily to equilibrium configurations. Wenzel was the first to deduce an expression for the apparent contact angle on rough substrates based on simplistic energy arguments, that could be verified in experiments with relatively large contact angles [7, 8, 9], but is nevertheless violated for the grooved substrates considered in the present study as demonstrated in the recent experiments of Chung et al. [10]. Axisymmetric droplet equilibria for substrates with concentric grooves were studied by Johnson & Dettre [11] and Huh & Mason [12] while Cox [13] generalized their work to include arbitrary, three-dimensional substrates. The last author in particular investigated the case of an infinite fluid wedge and was primarily concerned with the existence of multiple droplet equilibria. His work suggested that the hysteresis that can be induced by the substrate features would be higher for parallel-grooved, two-dimensional substrates. He also conjectured that contact line spreading normal to the grooves will be harder than spreading in all other directions and that it will exhibit a stick-slip behavior. Evidence for such behavior was reported in the exploratory experiments by Oliver et al. [14] and more recently by Chung et al. [10], mentioned earlier. The methodologies applied in the above studies were limited in scope however, principally because only equilibrium configurations were considered.
and quantitative statements about the dynamics were not made. As a matter of fact, dynamics has received much less attention compared to equilibrium, with a few notable exceptions which examined time-dependent problems numerically. We note, for example, the work of Gaskell et al. [15], who used the computationally advantageous precursor-film model to study droplet spreading over a three-dimensional topography by solving the long-wave approximation and the work of Gramlich et al. [16] who examined the motion of a single contact line without a precursor film over a two-dimensional topography by solving the Stokes equations.

Here we undertake a systematic investigation of both the equilibrium shapes and the motion of two-dimensional droplets over topographical substrates. The restriction to two dimensions implies that there are no transverse variations and the contact line is essentially treated as a set of two points. The inclusion of three-dimensional effects is much more involved, but we believe that our model is able to capture many of the qualitative features of the spreading behavior of a three-dimensional droplet. We shall hereinafter refer to a two-dimensional droplet simply as droplet.

Our starting point is the generalization of the work of Hocking on flat substrates [17] to topographical ones. We assume that substrate variations occur at lengthscales that are much longer than the slip length. We subsequently assess analytically the effects of the substrate topography on the motion of a droplet in the limit of $Ca \ll 1$, where $Ca$ is the capillary number, defined as

$$Ca = \frac{\mu U}{\sigma},$$

where $\mu$ is the fluid viscosity, $\sigma$ is the surface tension and $U$ is the (time-dependent) rate of spreading. Indeed, typical experimental configurations fall within this regime, but more importantly this assumption allows us to regard the droplet motion as quasi-static.

Assuming small angles everywhere, a single equation that describes the evolution of the droplet thickness is used, which is obtained via a long-wave expansion of the Stokes equations. We expect sharp variations in the droplet free-surface close to the contact points which suggest the presence of boundary layers there. We thus identify in §3 the bulk of the droplet as the outer region and the region near the contact lines as the inner regions. By asymptotically matching the two, we deduce the rate of spreading of the two moving fronts.

The equations obtained by matching are solved numerically for a number of test cases in §4. The dynamics of spreading, explored in more detail in §4.1, reveals that it is possible for the contact line to exhibit a stick-slip movement, especially during spreading on low-amplitude, short-wavelength substrates. On such substrates, a hysteresis-like effect is also observed and discussed briefly. By examining the phase-plane portrait of the two contact lines in §4.2, we analyze the existence of multiple equilibrium configurations and their nature as well as how these are affected by varying the substrate properties. A summary of our results is offered in §5.

2. Problem setup

We consider a droplet of a partially wetting fluid of density $\rho$, surface tension $\sigma$ and viscosity $\mu$ spreading on an arbitrary substrate. The cross-section of the droplet lies in the $x - z$ plane and is of infinite extent in the $y$–direction. The substrate is given by $z = \eta(x)$, and at time $t$ the profile of the free surface is located at $z = H(x, t) + \eta(x)$, where $H(x, t)$ corresponds to its thickness. The substrate is assumed to be a smooth function of $x$, so that there are no sharp edges that would prevent a clear definition of a contact line there [18]. Under the long-wave approximation [19] in which the slope of the droplet profile is assumed to be everywhere small we deduce a single equation for the droplet thickness:

$$\frac{3\mu}{\sigma} \partial_t H + \partial_x \left[ H^2 \left( H + 3\lambda \right) \partial_x^2 \left( H + \eta \right) \right] = 0,$$
where \( \bar{\lambda} \) corresponds to the slip coefficient, introduced to alleviate the non-integrable stress singularity that would otherwise appear at the contact points [20]. Equation (2) is made non-dimensional by writing

\[
x = Lx_s, \quad t = \frac{3\mu L}{\sigma \tan^{3}\alpha_s} t_s, \quad \bar{\lambda} = \frac{1}{3} L\lambda \tan \alpha_s, \quad \bar{\eta} = L\eta \tan \alpha_s \quad \text{and} \quad H = LH \tan \alpha_s,
\]

where \( \alpha_s \) is the static contact angle and \( L \) is a length scale defined by the droplet cross-sectional area, \( A \):

\[
L = \sqrt{\frac{A}{2\tan \alpha_s}}.
\]

Equation (2), written in non-dimensional form, becomes:

\[
\partial_t H + \partial_x \left[ H^2 (H + \lambda) \partial_x^3 (H + \eta) \right] = 0.
\]

For convenience we shall henceforth drop the stars in (5) and proceed using the non-dimensional variables. For a droplet with contact lines at \( x = a_\pm(t) \) such that \( a_-(t) \leq x \leq a_+(t) \), the boundary conditions at \( x = a_\pm(t) \) are

\[
H = 0 \quad \text{and} \quad \partial_x H = \mp \tan \theta_\pm,
\]

where the \( \theta_\pm \) are given by

\[
\tan \theta_\pm = \frac{1 + \tan \alpha_s^2 \eta_\pm^2}{1 \pm \tan \alpha_s^2 \eta_\pm^2},
\]

where we set \( \eta_\pm = \partial_x \eta |_{x = a_\pm} \). The last geometric conditions essentially fix the angles the free-surface makes with the substrate to their static value. Finally, the cross-sectional area of the droplet remains constant, leading to the global condition

\[
\int_{a_-}^{a_+} H \, dx = 2,
\]

with the chosen lengthscales. The solution to (5) depends on two parameters, the non-dimensional slip length, \( \lambda \), which is always small, and the contact angle, \( \alpha_s \). We thus expect to have a boundary layer in the vicinity of the droplet contact line, where an abrupt change in \( \partial_x H \) takes place to match the microscopic, static contact angle to the apparent contact angle in the bulk.

In what follows, we will examine the effects of the substrate profile \( \eta(x) \) on the spreading dynamics. In principle, \( \eta(x) \) can be arbitrary as long as it satisfies \( \partial_x \eta \leq \mathcal{O}(1) \). In addition, we require that the contact line is not trapped within a feature of the substrate. This can be avoided by appropriately choosing the substrate amplitude and by requiring the substrate variations to occur at a lengthscale that is much longer than the slip length.

### 3. Singular Perturbation Expansion

The problem is analytically tractable if we restrict our discussion to the limiting case \( Ca \ll 1 \) as noted in § 1 and as done in the earlier studies on flat substrates for both the slip [17] and constant-thickness precursor film [21] models. In this limit, surface tension forces dominate viscous ones. This implies that in a region sufficiently far from the contact line, the droplet shape is primarily affected by surface tension, allowing a different treatment of the governing equation in the bulk of the fluid (outer region) from the vicinity of the contact lines (inner regions). By properly matching the outer solution with the inner ones, we can obtain the velocities of the two moving fronts. Matching with a precursor-film inner region is discussed in [22, 23].
3.1. Outer region

For convenience, we transform the domain $a_- \leq x \leq a_+$ to $-1 \leq y \leq 1$, via the mapping:

$$x = \frac{1}{2} (a_+ - a_-) y + \frac{1}{2} (a_+ + a_-),$$

which allows us to write (5) for the outer region in the (non-moving) coordinate $y$ as:

$$\partial_t H - \frac{\dot{a}_+ (1 + y) + \dot{a}_- (1 - y)}{a_+ - a_-} \partial_y H + \frac{16}{(a_+ - a_-)^3} \partial_y [H^3 \partial_y^3 (H + \eta)] = 0,$$

where $\dot{a}_\pm = da_\pm /dt$ correspond to the spreading rates of the two contact lines at $x = a_\pm(t)$. In (10), we dropped $\lambda$ because slip is not significant in the outer region. The limit $Ca \ll 1$ is equivalent to $|\dot{a}_\pm| \ll 1$, which prompts us to introduce a quasi-static expansion for $H(x, t)$ in the form:

$$H(x, t) = H_0 (y, a_\pm) + H_1 (y, \dot{a}_\pm, a_\pm) + \cdots,$$

where the time dependence of $H(x, t)$ enters through $a_\pm(t)$ and their time derivatives. The task here is to approximate $H(x, t)$ as we approach the contact lines with the aim of asymptotically matching with the solution coming from the inner region. To leading order in $a_\pm$ we have:

$$\partial_y^3 (H_0 + \eta) = 0.$$

This equation is third order in $y$ and cannot, in general, satisfy all conditions (6) and (8). Since the contact angle boundary conditions apply at the contact lines, which are outside the domain of the outer solution, we use (6) and (8) to obtain $H_0(y, t)$:

$$H_0 = \frac{3}{2} \left[ \frac{2}{a_+ - a_-} + \bar{\eta} - \frac{1}{2} (\eta_+ + \eta_-) \right] (1 - y^2) + \frac{1}{2} [\eta_+ (1 + y) + \eta_- (1 - y)]$$

$$- \eta \left( \frac{1}{2} (a_+ - a_-) y + \frac{1}{2} (a_+ + a_-) \right),$$

where $\eta_\pm = \eta(a_\pm)$ and $\bar{\eta} = \int_{a_-}^{a_+} \eta(\xi) \, d\xi / (a_+ - a_-)$. The term $\partial H_0 / \partial t$ is approximated as $\partial H_0 / \partial t = \dot{a}_+ \partial H_0 / \partial a_+ + \dot{a}_- \partial H_0 / \partial a_-$. And appears in the equation for the next term, $H_1 (y, t)$, which is solved subject to homogeneous boundary conditions, namely

$$H_1 (\pm 1) = 0 \quad \text{and} \quad \int_{-1}^{+1} H_1 \, dy = 0.$$

Finding $H_1$ cannot be done analytically for arbitrary $\eta(x)$, but what we are interested in is the leading-order behavior of $H_1$ as $y \to \pm 1$. The third derivative of $H_1$ is singular at $y = \pm 1$ and behaves like

$$\partial_y^3 H_1 = \frac{a_+ - a_-}{2 \phi_\pm^2 (1 \mp y)^2} \dot{a}_\pm + O \left( (1 \mp y)^{-1} \right),$$

where

$$\phi_\pm = \frac{2}{a_+ - a_-} \left[ \frac{6}{a_+ - a_-} + 3 \bar{\eta} - 2 \eta_\pm - \eta_\mp \pm 1/2 \eta_\mp'(a_+ - a_-) \right].$$

Here $\phi_\pm$ correspond to the apparent contact angles obtained by evaluating $\partial_y H_0$ at $x = a_\pm$. In the limit of $\eta \ll H$, $\phi_\pm$ can be viewed as mesoscopic angles and the droplet spreads with an apparent angle, $\psi = 12 / (a_+ - a_-)^2$. Successive integrations yield the leading order behavior for $\partial_y H_1$ as the contact lines are approached:

$$\partial_y H_1 \sim - \dot{a}_\pm \frac{a_+ - a_-}{2 \phi_\pm^2} \left[ \ln \frac{e}{2} (1 \mp y) + \beta_\pm \right] \text{ as } y \to \pm 1,$$

where

$$\beta_\pm = \frac{1}{2} \left( \frac{a_+ - a_-}{a_+ - a_-} \right) \ln \frac{e}{2} (1 \mp y).$$
where the constants $\beta_{\pm}$ are found by appropriately integrating the equation for $H_1$ and making use of the conditions (14) to find:

$$\beta_{\pm} = \int_{-1}^{1} \frac{1}{1 + y} \left \{ 1 - \frac{(a_+ - a_-)^3 \phi_{\pm}^2}{128 \hat{a}_{\pm} H_0^3} \left [ (\hat{a}_+ \phi_+ - \hat{a}_- \phi_-) y + \hat{a}_+ \phi_+ + \hat{a}_- \phi_- \right ] \right \} \, dy. \tag{17}$$

By writing (13) and (16) in terms of $x$ and combining them, we find the slope of $H$ as $x \to a_{\pm}(t)$:

$$\mp \partial_x H \sim \phi_{\pm} \pm \frac{\hat{a}_{\pm}}{\phi_{\pm}^2} \left [ \ln \left ( e^{\pm a_{\pm} / a_+} \mp x \right ) + \beta_{\pm} \right ]. \tag{18}$$

These asymptotic expansions will ultimately be matched asymptotically with the inner solutions to be derived in the vicinity of the contact lines. For the expansions (18) to be valid, we require that matching occurs sufficiently close to the contact lines, so that the linear term of $\partial_x H$ is of higher order than the logarithmic term, which in turn is of higher order than the constant term in the expansion.

3.2. Inner region

The inability of the outer solution to satisfy the constant contact-angle boundary condition near the contact lines, necessitates the presence of inner regions where slip is significant and the above condition is exactly satisfied. To treat the inner region near $x = a_{\pm}$ for which $x - a_{\pm} = O(\lambda)$, we make the change of coordinates

$$H = \lambda \Phi \quad \text{and} \quad x = a_{\pm} \mp \lambda \xi / \theta_{\pm}, \tag{19}$$

which transforms (5) to

$$\lambda \partial_t \Phi \pm \hat{a}_{\pm} \theta_{\pm} \partial_x \Phi + \theta_{\pm}^2 \left [ \Phi^2 (\Phi + 1) \partial_{\xi}^2 \Phi \right ] = 0, \tag{20}$$

where it is assumed that $\lambda |\xi \partial \theta_{\pm} / \partial a_{\pm}| \ll 1$. Note also that the contribution $\partial_x \left [ H^2 (H + \lambda) \partial_{\xi}^2 \eta \right ]$ in (5) originating from the substrate is neglected, as we have made the assumption that the variations in $\eta(x)$ take place at lengthscales that are much longer than the slip length, and hence the substrate is locally treated as being flat. However, the substrate topography still affects the inner region dynamics through $\theta_{\pm}$ in equation (7), which depends on $\partial_x \eta$. These contributions are generally small, of $O\left ( \tan^2 \alpha_s \right )$, but are included in our model to exactly force the contact angle boundary condition.

Since we have assumed that the droplet spreads slowly, we can write

$$\Phi = \Phi_0 + \Phi_1 + \cdots \tag{21}$$

with $\Phi_0 = \xi$ and $\Phi_0 \gg \Phi_1$. The equation for $\Phi_1$ thus becomes:

$$\partial_{\xi}^2 \Phi_1 = \mp \frac{\hat{a}_{\pm} / \theta_{\pm}^3}{\xi (\xi + 1)}, \tag{22}$$

which is solved subject to the conditions that $\Phi_1 = \partial_{\xi} \Phi_1 = 0$ at $\xi = 0$ and $\Phi_1 / \xi^2 \to 0$ as $\xi \to \infty$ to get:

$$\partial_{\xi} \Phi_1 = \pm \frac{\hat{a}_{\pm}}{\theta_{\pm}^3} \left [ (\xi + 1) \ln (\xi + 1) - \xi \ln \xi \right ] \tag{23}$$
The asymptotic behavior of $\partial_{\xi} \Phi$ as $\xi \to +\infty$ is readily obtained as:

$$\partial_{\xi} \Phi \sim 1 \pm \frac{\dot{a}_\pm}{\theta^3_\pm} (\ln \xi + 1) \quad \text{as} \quad \xi \to +\infty.$$  \hfill (24)

which, in terms of the outer variables, is expressed as:

$$\mp \partial_x H \sim \theta_\pm \pm \frac{\dot{a}_\pm}{\theta^3_\pm} \left[ \ln \left( \frac{\theta_\pm (\pm a_\pm \mp x)}{\lambda} \right) + 1 \right] \quad \text{as} \quad \pm a_\pm \mp x \to \infty.$$  \hfill (25)

### 3.3. Asymptotic matching

Apparently the logarithmic terms in the inner (25) and outer (18) regions do not generally match and therefore an intermediate region would be required to properly continue the matching procedure, as in the flat substrate case [17]. We notice, however, that matching can actually be successful if it is done for $(\partial_x H)^3$ instead of $\partial_x H$. Thus the sole purpose of an intermediate region is precisely to justify matching $\partial_x H$ and for the sake of brevity it is omitted in the present treatment. Hence, considering the cube of (18) and (25), the logarithmic terms match provided that the matching is carried within the overlap region of validity of the two asymptotic expansions. Matching the constant terms then in the asymptotic expansions yields:

$$\phi^3_\pm - \theta^3_\pm = \pm 3 \dot{a}_\pm \left[ \ln \left( \frac{\theta_\pm (a_+ - a_-)}{\lambda} \right) - \beta_\pm \right].$$  \hfill (26)

Hence, the order of magnitude of the speed of the moving front is $|\dot{a}_\pm| \sim \mathcal{O}(1/|\ln \lambda|)$. Equations (26) constitute a system of integro-differential equations for $a_\pm(t)$, which are coupled through $\beta_\pm$. Explicit expressions for $\dot{a}_\pm$ can be found after term re-arrangement in (26) and solving the resulting system:

$$\dot{a}_\pm = \pm \frac{\delta_\pm I_\pm + \delta_\pm \phi_\pm I_0}{I_+ I_- - \phi_+ \phi_- I_0^2};$$  \hfill (27)

where

$$\delta_\pm = \frac{1}{3} \left( \phi^3_\pm - \theta^3_\pm \right),$$

$$I_\pm = \ln \left[ \theta_\pm \frac{a_+ - a_-}{\lambda} \right] + \int_{-1}^{1} \frac{1}{1 + y} \left[ \frac{\phi^3_\pm (a_+ - a_-)^3 (1 - y^2)^4}{128 H_0^3 (1 + y)} - 1 \right] \, dy,$$

$$I_0 = \frac{\phi_+ \phi_- (a_+ - a_-)^3}{128} \int_{-1}^{1} \frac{(1 - y^2)^3}{H_0^3} \, dy.$$

Equations (27) reveal an interesting interplay between the dynamics of each contact line. Equilibrium not only depends on whether the apparent contact angle becomes equal to the static one ($\delta_\pm = 0$), but also on how fast the other contact line moves. In fact, as our results indicate asymmetric spreading is almost always accompanied by a recession of one of the contact lines before the droplet reaches equilibrium.

### 4. Results

#### 4.1. Spreading dynamics

We now proceed with presenting a few typical solutions to (27), aiming to illustrate the essential features of the spreading dynamics and to gain further insight into the effects of substrate heterogeneities. Since the overall picture is not drastically altered by variations in the slip length and the static contact angle, in all cases presented hereinafter, we fix $\lambda = 10^{-4}$ and $\alpha_s = 5^\circ$ using $a_\pm(0) = \pm 1$ as initial conditions for time integration.
We begin by considering the substrate $\eta(x) = (2 \sin x + \sin 2x + 2 \cos x)/10$, which corresponds to a $2\pi$-periodic series of bumps. In Fig. 1(a), we present the evolution of the moving fronts as a function of time. At first, both fronts advance, but at a later stage the right front begins to recede as the droplet slides downhill towards equilibrium (see Fig. 2, where we show the outer solution, (13), at different times). As it turns out, this brief recession of the contact line is always present unless spreading is perfectly symmetric. This behavior can be alternatively observed by looking at the evolution of the ratio of $\phi_\pm/\theta_\pm$ evaluated at the two contact lines, as shown in Fig. 1(b). As our analysis suggests, contact line advancement occurs provided that $\phi_\pm > \theta_\pm$. Even though the right front recedes for a relatively long period of time, it does so very slowly.

The moving contact lines may exhibit a stick-slip-type behavior while they move along the substrate features as Cox conjectured [13]. To illustrate this effect, we consider the small-amplitude, periodic substrate given by $\eta(x) = 0.003 \sin 80x + 0.005 \cos 40x$. Figure 3(a) shows the location of the two moving fronts as a function of time. For comparison, we also plot the contact line location for ideally flat substrates. Even though at early times the droplet tends to spread nearly symmetrically following the overall trend of an ideally flat substrate, the contact line velocities are highly oscillatory as can be seen in Fig. 3(b), where we plot their time evolution. Sticking and slipping is particularly evident for the left contact line, as the staircase-like curve for $a_-(t)$ suggests. We can therefore conclude that substrate heterogeneities tend to have a greater impact on the dynamics as the droplet approaches equilibrium (see Fig. 4(a)).
Figure 3. Spreading on a substrate $\eta(x) = 0.003 \sin 80x + 0.005 \cos 40x$ with $a_\pm(0) = \pm 1$, $\lambda = 10^{-4}$ and $\alpha_s = 5^\circ$. (a) Evolution of moving fronts’ locations (solid and dashed lines) demonstrating stick-slip-type behavior; the dotted curve corresponds to the contact line location for $\eta(x) = 0$. (b) The corresponding velocities of the contact points.

Figure 4. Droplet profiles for spreading on a substrate (a) $\eta(x) = 0.003 \sin 80x + 0.005 \cos 40x$ and (b) $\eta(x) = 0.005 \cos 40x$, at times $t = 0$ (dashed curve) and $t = \infty$ (solid curve). The dotted curve corresponds to the equivalent droplet equilibrium profile had the substrate been flat illustrating the substrate-induced hysteresis effect upon comparison with the solid curve.

is also surprising is the fact that the right contact line is nearly stationary while sticking and slipping occurs. This example suggests that the temporary pinning of the contact line, reported in experimental studies [2], may be attributed at least in part to the details of the substrate topography. Of course other surface features, such as chemical defects/heterogeneities can also be responsible.

As the preceding example suggests, even small-amplitude spatial heterogeneities can dramatically alter the spreading dynamics. In Fig. 4(a) we show the equilibrium attained for the example in the previous paragraph, where we can see an appreciable shift of the droplet to the left. Contrary to the flat-substrate case, we also notice that it is possible for the apparent contact angle at equilibrium to be quite different from the static contact angle. Further evidence for substrate-induced hysteresis can be found in Fig. 4(b), where we plot the equilibrium attained while spreading symmetrically on the substrate $\eta(x) = 0.005 \cos 40x$. This observation prompts us to investigate under what conditions the hysteresis-like effect induced by topographical substrate variations is more prominent. As noted earlier, when $|\eta(x)| \ll 1$
Figure 5. The \((a_+, -a_-)\) phase plane for \(\eta(x) = (\cos 6x + \sin 3x)/12\). There are multiple droplet equilibria, that may correspond to stable nodes (solid circles), unstable nodes (open circles) or saddle points (crossed circles). For comparison the dotted line shows the line equilibria for ideally flat substrates. Solid and dashed curves correspond to stable and unstable saddle point manifolds, respectively, which delimit the basins of attraction of the stable fixed points. Since not all initial conditions are permissible, the dash-dotted line is used to separate the physical and non-physical (free surface touches the substrate at a point different to a contact one) parts of the \((a_+, -a_-)\) phase plane. The shaded region corresponds to the region of the phase plane that is accessible when both contact lines advance. The translational invariance due to the substrate periodicity implies a finite number of equilibria, namely 5 stable nodes (denoted by A–E), 6 saddle points and 1 unstable node.

the apparent contact angle for both contact lines is \(\psi = 12/(a_+ - a_-)^2\), which at equilibrium will be different from the actual angle the substrate makes with the droplet free surface by an amount:

\[
\Delta \phi_{\pm} = \phi_{\pm} - \psi = \frac{2}{a_+ - a_-} \left[ 3\bar{\eta} - (2\eta_{\pm} + \eta_+) \right] \pm \frac{1}{2} \eta_{\pm} (a_+ - a_-). \tag{28}
\]

Given that the amplitude of the features is small, this substrate-induced hysteresis in the apparent contact angle can be appreciable when variations in \(\eta(x)\) are relatively ‘fast’ but \(\partial_x \eta \lesssim 1\), to be in accord with the assumptions of our long-wave expansion.

4.2. Phase-plane analysis

Our expectation that multiple equilibria exist, motivates the investigation of the droplet spreading dynamics on the phase plane. When the droplet reaches equilibrium, \(\dot{a}_\pm = 0\), so that (18) becomes \(\phi_{\pm eq} = \theta_{\pm eq}\), where we use \(a_{\pm eq}\) to denote the equilibrium positions of the
Figure 6. Equilibrium droplet profiles corresponding to the stable equilibria A–E of Fig. 5. Equilibrium shapes B, C and E enhance wetting, but their basins of attraction are significantly smaller than shapes A and D.

contact lines at \(x = a_\pm(t)\). Spreading on a flat substrate is translationally invariant, with \(a_+\) and \(a_-\) related through

\[
a_- = a_+ - 2\sqrt{3},
\]

so that there exists a continuum of equilibrium configurations. The introduction of spatial heterogeneities drastically changes the attainable steady states. To demonstrate this, consider the substrate \(\eta(x) = (\cos 6x + \sin 3x)/12\). Now the continuum of equilibria given by (29) is destroyed and countably infinite new equilibrium positions emerge. In Fig. 5 we show the equilibrium points on the \(a_+ - a_-\) phase plane along with the direction field that corresponds to solutions to (27). Our analysis reveals that these equilibria can be either stable or unstable nodes or saddle points, but they do not necessarily correspond to different states. The periodicity of the substrate considered has an inherent translational invariance with respect to the wavelength of the topographical feature. Accounting for the substrate periodicity, we have in fact five distinct stable equilibria, shown as points A–E on the phase plane. Three of these are symmetric about the topographical features (points A, B and D). All equilibrium shapes corresponding to these contact line locations are given in Fig. 6. It is important to note that not all \(a_\pm(0)\) would yield physically meaningful solutions to (27). In fact, a rather large part of the phase plane is excluded, since we require that \(a_+ > a_-\) and that the droplet free surface does not touch or cross the topographical features (irregularly-shaped domain in the lower right of Fig. 5). On the remaining phase-plane, we see that the equilibrium shapes A and D have significantly larger basins of attraction than the other three equilibria. For these two shapes it is readily seen that the droplet wets the droplet less than the flat-case equilibrium. Wetting-enhancing equilibria, B, C and E, are more accessible through contact line recession than spreading. In principle however for the majority of permissible initial conditions both fronts are advancing at early times (shaded region in Fig. 5).

5. Conclusions
We have developed a model for describing the surface-tension dominated motion of a partially wetting two-dimensional droplet over spatially heterogeneous substrates in the Stokes-flow.
regime. Assuming small contact angles that remain constant and equal to their static value along the contact lines and that substrate variations occur over length scales that are much larger than the slip length, we obtained a single equation for the evolution of the droplet thickness and its contact lines obtained via a long-wave approximation of the Stokes equations. The force singularity at the contact lines was alleviated through the Navier slip condition. For small capillary numbers, the droplet approaches equilibrium quasi-statically, with the time-dependence entering the problem through the location of the contact lines. This allowed us to regard the dynamics as a perturbation problem in terms of the spreading rates of the two droplet fronts. By matching the flow in the vicinity of the contact lines with the flow in the bulk of the droplet, we were able to obtain a set of two coupled integro-differential equations for the location of the droplet fronts.

As the droplet moves along the topographical features, the contact lines may exhibit a stick-slip-type behavior, especially if the characteristic wavelength of these features is small. Such behavior is more readily observed as the droplet approaches equilibrium, where the forcing of the substrate can more easily influence the slowly moving droplet. This effect should not be confused with the sticking and slipping that may occur when chemical defects are present. What this calculation suggests, however, is that the substrate features may amplify this effect. We have also demonstrated via a phase-plane analysis that substrate heterogeneities allow for the existence of multiple equilibrium droplet configurations. The complexity of the phase plane, the number of equilibria and their nature are highly dependent on the density and height of the topographical features. As a matter of fact, we observe that a spatially heterogeneous substrate can induce a hysteresis-like effect, which can become appreciable for sufficiently ‘rough’ substrates.

Even though the inclusion of more realistic, three-dimensional substrates would yield results that can be directly compared with existing experiments, we believe that many of the essential features can be adequately studied with our model. It is thus hoped, that through the formalism developed in the present study, other related problems pertaining to the dynamics of droplet spreading can be addressed, such as the effects of random substrates on the statistics of the location of the contact lines. We shall examine this and related issues in future studies.

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