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Contact-line instability of liquids spreading on top of rotating substrates

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ABSTRACT

A (film or) droplet of viscous liquid spreads isothermally on a smooth horizontal solid surface. The lubrication approximation is used to study the linear stability of thin (films or) droplets, subject to capillary, gravitational, and centrifugal forces, and a variety of contact-angle-versus-speed conditions. All equations are derived for plane spreading films and rotationally-symmetric spreading droplets, while the discussion of the results is carried out for the droplets. It is found that in general two types of two-dimensional base states develop. Early on there is a simple convex contour and later a contour with a pronounced capillary ridge near the contact line. While the convex contour remains stable, the capillary-ridge contour becomes unstable with regard to disturbances, which are periodic in the lateral direction. As the contact line advances in time, this instability involves a transition from two-dimensional to three-dimensional spreading, whereas modes with increasing wave numbers become successively unstable. The onset of the instability is controlled by gravitational, centrifugal, and capillary forces, whereas gravitational and capillary forces tend to stabilize and centrifugal forces tend to destabilize the system. For partially-wetting systems, the neutral stability appears to be not affected by the static advancing contact angle, though the growth rates are modified.

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1. Introduction

The spreading of liquids on solids under various conditions has been investigated in literature to a reasonable extent. Extensive reviews have been presented by de Gennes [1], or more recently, by Bonn et al. [2]. One important aspect is the physics of the dynamic contact line. The characteristics of such dynamic contact lines and their treatment within the frame of continuum mechanics have been discussed in detail by Dussan V. [3]. In the cited reviews, a further aspect, namely the effect of auxiliary forces onto the spreading liquid, plays a minor role. Even though gravitational forces are addressed to some extent, centrifugal forces due to a rotating substrate are rarely within the focus of attention. However, it is the effect of centrifugal forces which relates a spreading liquid layer to the early stage of spin coating, during which the contact line moves to the edge of the substrate. The spin-coating process is widely engaged in industry to coat advanced electronic or optical devices with solid layers, which are achieved via liquid spreading and subsequent solvent evaporation (cf. Larson and Rehg [4] for a review). Finally, the issue of the stability of an advancing liquid front on a solid is not yet fully resolved. Experiments sketched below in detail indicate that for driven contact lines a fingering instability may occur. Hereby, the driving of the contact line may be due to the wall-tangential components of auxiliary forces. Prominent examples are gravitational forces during liquid spreading down an inclined plate, centrifugal forces during liquid spreading on a rotating plate, or air-jet blowing during liquid spreading on horizontal plates. The analogy of these three methods of external driving has been pointed out e.g. by Troian et al. [5] or Moriarty et al. [6]. Further, most examples have in common that the spreading liquid flow usually is characterized by a small Reynolds number, such that the Stokes equations may be used as the basis for a theoretical treatment (cf. Goodwin and Homsy [7]).

An early theoretical investigation into a liquid layer on a rotating solid plate has been conducted by Emslie et al. [8]. For his treatment he assumes that a thin Newtonian liquid layer exists on the rotating plate, which is rotationally-symmetric. Without the need to deal with a (moving) contact line, his evolution equation involves simply a balance of viscous and centrifugal forces. He finds that initially uniform liquid layers remain uniform in the presence of the rotation, and that initially non-uniform liquid layers tend towards uniformity due to the rotation. Davis [9] is one of the first authors who concentrates on the stability of moving contact lines. For the specific case of a so-called rivulet, i.e. a narrow stream of liquid on a solid, he poses three different conditions for the contact-line dynamics on both sides of the rivulet, namely a fixed contact

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line, a moving contact line with fixed contact angle, and a moving contact line with a smoothly varying contact angle. Engaging an energy method, he arrives at a damped linear harmonic-oscillator equation and derives stability conditions for all three contact-line conditions. He concludes that for base states with dynamic contact lines, the energy method appears to be inappropriate.

Huppert [10] presents experimental results for the spreading of a viscous current down an inclined plate. In particular, he finds a fingering instability of the contact line and derives an expression for the wavelength of the disturbances, which appears to be independent of both viscosity and contact-line characteristics. Schwartz [11] infers numerical solutions to an evolution equation, valid for the viscous flow down an inclined plate with both gravity and surface tension acting. Invoking the lubrication approximation, he finds for a contact angle of zero, i.e. for perfect wetting, interfaces which feature the formation of fingers. Troian et al. [5] employ a two-zone model, with an outer zone of constant liquid thickness and an inner capillary zone near the moving contact line, in conjunction with the lubrication approximation. From a linear stability analysis, they conclude that thin films with small contact angles, driven by external body forces, can be unstable to periodic disturbances and, hence, to the formation of fingers. Likewise based on a two-zone model, Moriarty et al. [6] construct a matched-asymptotic solution for a thin liquid film draining down a vertical wall. These authors further extend their theoretical investigations towards the spinning film on a rotating horizontal substrate and towards the blowing of an air jet onto a spreading film. In all cases they find time-dependent interface profiles, which develop a steep capillary ridge near the front. The stability question is not addressed by these authors. Bertozzi and Brenner [12], also on the basis of a two-zone model, revisit the stability of the viscous current on an inclined plate, as first discussed by Huppert [10]. In addition to the (driving) wall-tangential contribution of gravity, they account for the wall-normal contribution and infer that below a critical inclination angle the base state appears to be linearly stable. Further, from numerical simulations, they establish that there is significant growth of microscopic-scale perturbations at the contact line on a transient time scale.

An early experimental confirmation of the finger formation at droplets, spreading on rotating substrates, is presented by Melo et al. [13]. These authors capture the evolution of the droplet footprint in time and find, at first, axis-symmetric spreading, followed by the formation of a capillary ridge near the contact line, and finally, the appearance and growth of disturbances, which are periodic in the circumferential direction. Further, a comparison with a two-zone model is presented, featuring reasonably-good agreement. In two articles, Fraysse and Homsy [14] and Spaid and Homsy [15] address experimentally the spreading and the instability of Newtonian and non-Newtonian liquids on rotating substrates. By registering translucent light with a CCD camera from above, the authors capture the contact line and, from absorption due to a dye, infer droplet profiles, both as a function of time. The authors report that the most unstable wavelength appears to be independent of both the droplet size and the rotation speed, however, it depends on the wetting characteristics. Further, no significant difference has been found between a (non-Newtonian) Boger fluid and its (Newtonian) solvent. This is attributed to the small Weissenberg number within these experiments. For larger Weissenberg numbers, though, the non-Newtonian fluid causes a delay of the instability and a reduced growth rate of the disturbances if compared to its (Newtonian) solvent. Moreover, Spaid and Homsy [16] extend the stability analysis of Troian et al. [5] to non-Newtonian fluids. In detail they analyze the stability of a straight capillary ridge based on a two-zone model with either a precursor film or slip at the front. They sketch the mechanism of this instability, namely that thick regions of the capillary ridge are more affected by the volumetric force and less affected by the wall presence. Hence, these regions advance faster and the flow is redirected into these fingers. They further find that the instability is little affected by the details of the contact-line model and the contact angle. However, the viscoelastic stabilization from the experiments is confirmed by their stability analysis.

McKinley et al. [17] theoretically investigate the spreading of an incompressible Newtonian liquid droplet attached to a horizontal plate and subject to air-jet blowing. They rely on the conservation equations in the lubrication limit, model the dynamic contact line by means of the generalized law of Tanner, and invoke the limit of small capillary numbers to infer an evolution equation and quasi-steady solutions to the problem. They discuss spreading droplets without gravity, and with gravity for sessile and pendant droplets, all for a partially-wetting system. They find simple convex or concave droplets, and droplets which develop a capillary ridge near the front, or even open in the centre to form an annular ring of liquid. Further, they analyze the stability of equilibrium solutions with regard to small axis-symmetric disturbances and find, independent of gravity, that the non-annular equilibrium droplets remain stable. However, annular droplets without gravity appear to be unstable. McKinley and Wilson [18] extend this stability analysis to non-axis-symmetric disturbances and to finite capillary numbers. For neglected gravity, they find all equilibrium solutions to be unstable, both for the quasi-steady limit of vanishing capillary numbers and for finite capillary numbers. This result holds for non-annular and annular droplets. Wilson et al. [19] engage a similar framework to consider the axis-symmetric spreading of droplets on top of a rotating substrate. They infer analytical solutions for vanishing and small surface tension, and numerical solutions for finite surface tension, while gravity is neglected. Their solutions for finite surface tension recover the development of a capillary ridge near the moving contact line and prove to be in agreement with the experimental findings of Fraysse and Homsy [14] and Spaid and Homsy [15], obtained for Newtonian fluids prior to the onset of the fingering instability. The stability question is not addressed. Schwartz and Roy [20], based on numerical solutions of the evolution equation, lubrication approximation, and (long-range) molecular forces, focus on the spreading of a partially-wetting viscous liquid on a rotating substrate. By means of an energy method, the stability of axis-symmetric equilibrium solutions is in particular addressed. They find all equilibrium solutions to be unstable with respect to the off-centre displacement mode, while higher modes remain stable until the rotation speed exceeds some critical value. Further, the authors report that a comparison with the experimental results of Fraysse and Homsy [14] for the Newtonian solvent appears to be favourable.

In more recent experiments, Holloway et al. [21] study the spreading of silicone oil droplets on rotating substrates. The authors engage shadowgraphy to capture the footprint of the droplets in time and derive spreading laws for axis-symmetric spreading prior to the onset of the instability. Further, growth rates for various modes of the fingering instability are presented. The authors report that the number of fingers increases with both the rotation speed and the drop volume. Chou et al. [22] investigate the spreading of various silicone oils on a rotating silicon wafer by means of two CCD cameras, directed from the top and the side onto the droplet. From these images they infer the droplet radius and estimate the (dynamic) contact angle. The authors report a crucial influence of the dynamic contact angle onto the fingering instability and find contact angles of up to 150°, depending on the rotation speed. Further, the critical radius for the onset of the fingering instability is found to be a function of a modified rotational Bond number. Finally, Mukhopadhyay and Behringer [23] engage an interferometric technique to capture

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