# ELASTOHYDRODYNAMIC SOLID-LIQUID-SOLID DEWETTING

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The presence and stability of thin liquid films between two solid surfaces is of great scientific and technological relevance for many systems and applications. For minimizing wear in bearings, a stable liquid film is desirable. To prevent aquaplaning on wet roads, the liquid film should be removed as efficiently as possible to maximize traction. Similarly, for residue-free nanoimprint lithography, a swift and complete removal of intervening liquid is required. The strength of adhesion between organic or polymeric surfaces is considerably reduced by the presence of a water film, as the Hamaker constant for the interaction of e.g. two polystyrene or poly-tetra-fluoro-ethylene halfspaces across vacuum is 7 to 10 times higher than across an ultrathin water layer.

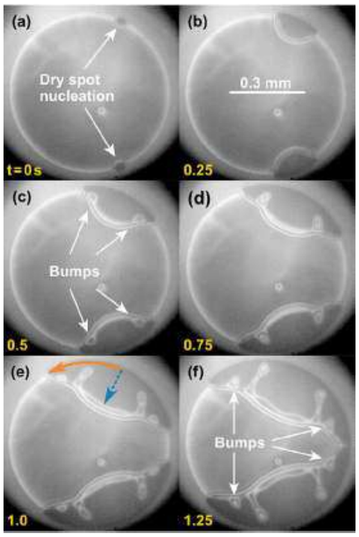

Fig. 1: (a) Simultaneous dry-spot nucleation in two locations on the perimeter of the contact spot. (b)–(f) The expansion rate of the dry spots is highly anisotropic and significantly faster along the edge of the contact spot [4].

Roberts and Tabor reported the spontaneous collapse of thin liquid films confined between a rubber surface and an SiO2 lens for film thicknesses below 40 nm in 1968. Brochard-Wyart and de Gennes were the first to present an analytical model for the dewetting of water films between a rigid solid and a rubber [1]. By balancing the surface energy change with the viscous dissipation in the liquid they derived the scaling law for the growth rate of an axisymmetric dry spot. Martin and Brochard-Wyart presented experiments that confirmed the predicted scaling laws [2]. Persson et al. additionally considered the non-uniformity of the pressure distribution inside the contact area [3].

Brochard et al. and Persson et al. focused on axisymmetric dewetting, which is a rather rare occurrence and needed to be induced intentionally by artificial defects or protrusions on one of the confining solids. In contrast, we conducted systematic experiments of non-axisymmetric dewetting. An elastomeric half-sphere is pressed onto a flat layer of the same material with liquid confined between them.

Due to the applied pressure, the liquid film thickness h decreases in time. After h has become sufficiently low, dry-spot nucleation occurs at regions of minimum film thickness, usually at the rim of the contact spot. Figure 1 illustrates a fortuitous example, where two dry-spots nucleated almost simultaneously at different locations around the rim. It can be seen that the shape evolution proceeds in an essentially mirror-symmetric fashion, which proves that the dewetting process is governed by hydrodynamic effects and is not obscured by random surface defects and heterogeneities. The arrows in Fig. 1(e) indicate that the dewetting speed is highly anisotropic and is higher along the perimeter than radially inwards. Moreover, the receding liquid rim does not remain flat and smooth but develops bumps that will eventually break-up into droplets. This instability is more pronounced for thinner films as seen in Fig. 2, where only a single dry spot nucleation took place. After their formation, these droplets are subsequently propelled towards the edge of the contact spot by the contact pressure gradient. The contact pressure is highest in the centre of the contact spot and decays towards its edge.

Fascinatingly, the coalescence process of a radially moving droplets with the bulk liquid outside of the contact spot can proceed in a continuous or discontinuous fashion. Fig. 3(a-i) shows an example of a partial, discontinuous coalescence. The liquid bridge connecting the droplet and outer liquid displays a non-monotonic behaviour. It initially grows [Fig. 3(b) and (c)] then shrinks [Fig. 3(d)] and eventually disintegrates [Fig. 3(e)]. This cascading behaviour can occur multiple times during the lifetime of a droplet. Up to nine consecutive cascades were observed for a single droplet. Figure 3(j) compares the time history of the droplet footprint area of a non-cascading droplet and one that cascades five times. The latter shows a pronounced staircase-like morphology with a relatively uniform area reduction ratio An /An-1.

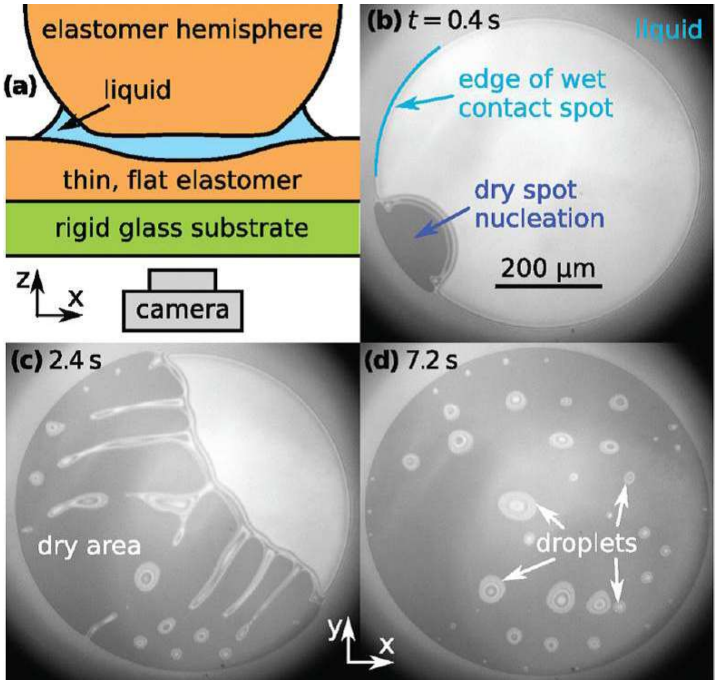

Fig. 2: (a) Side-view sketch of the experimental geometry. (b–d) Microscope images of dry spot nucleation and dewetting in the wet contact zone between an elastic half-sphere and a flat elastic layer. Frequently droplets are created due to an instability of the dewetting rim. (e) Microscope images of a droplet moving towards the edge of the contact spot [5].

The physical mechanism that causes the partial coalescence in this system is currently unclear, as is the reason why some droplets coalesce in a continuous fashion and others do not. Partial coalescence of spherical droplets at air-water interfaces is related to the cushioning effect of the gas interlayer and capillary-inertial effects, all of which play no role in our system.

We developed a theoretical model to simulate soft elastohydrodynamic lubrication phenomena and the dewetting of thin films between soft elastic surfaces numerically. The model combines the Reynolds equation for thin liquid film flow and linear elasticity accounting for the stresses and deformations of the soft elastomeric components. The three-dimensional implementation allows us to study non-axisymmetric geometries. The elastic deformation in the limit of slow deformation and small strain is governed by the Cauchy momentum equation for a stationary system, assuming the elastic material properties to be linear, non-dissipative, isotropic and homogeneous. The partial wettability is implemented in the elastohydrodynamic framework by means of the concept of disjoining pressure Π, for which an empirical relation based on two power-law terms was used.

We successfully validated our model using the available analytical models for axisymmetric dewetting. The model successfully predicts the effect of periodic surface topography on the shape of the dry spot [6]. Moreover, the model reproduces the morphology of the dewetting spots as well as their anisotropic growth dynamics shown in Fig. 1 very well [4]. The numerical simulations can also reproduce the shape and escape dynamics of non-cascading droplets in Fig. 2 qualitatively well [5].

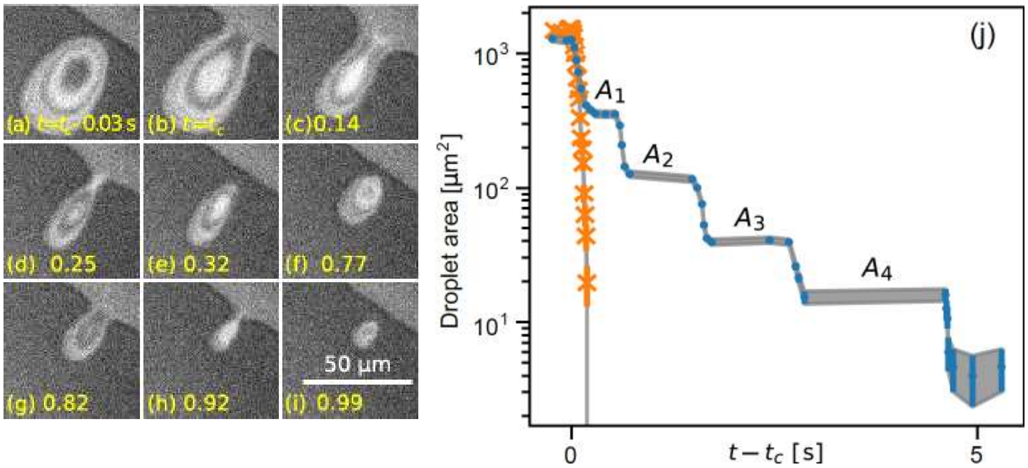

Fig. 3: (a-i) A droplet undergoing a discontinuous coalescence with the bulk liquid outside of the contact spot. The indicated time increments are given relative to frame (b) where coalescence started. The scale bar in (i) applies to all images. (j) Droplet footprint area as a function of time. The blue circles correspond to the data shown (a–i). The orange crosses refer to a droplet undergoing a continuous, non-cascading coalescence [5].

Droplets that undergo a coalescence cascade tend to move much slower than droplets that merge in a single coalescence event. Furthermore, the speed of motion of the cascading droplets is not proportional to the local pressure gradient. Using numerical simulations, we have investigated surface roughness and spatial modulations of Young's modulus and the spreading parameter as potential mechanisms that could slow the droplets down. As for the latter two, we generally found that for experimentally conceivable parameter variations, the reduction in contact line speed was far less than two orders of magnitude. In contrast, surface roughness induced a speed reduction comparable with the experimental observations and is thus a viable candidate for the responsible mechanism.

Additional details about the results that were briefly introduced above can be found in references [4-6].

### REFERENCES

1. Brochard-Wyart, F. and Gennes, P.G.D. (1994) Dewetting of a water film between a solid and a rubber. J. Phys.: Condens. Matter 6, A9.
2. Martin, P. and Brochard-Wyart, F. (1998) Dewetting at soft interfaces. Phys. Rev. Lett. 80, 3296.
3. Persson, B.N.J., Volokitin, A.I. and Tosatti, E. (2003) Role of the external pressure on the dewetting of soft interfaces. Eur. Phys. J. E 11, 409.
4. Chudak, M. K., Kwaks, J.S., Snoeijer, J.H. and Darhuber, A.A. (2020) Non-axisymmetric elastohydrodynamic solid-liquid-solid dewetting: Experiments and numerical modelling. Eur. Phys. J. E 43, 2.
5. Chudak, M. K., Kwaks, J.S., Snoeijer, J.H. and Darhuber, A.A. (2020) Escape dynamics of liquid droplets confined between soft interfaces: non-inertial coalescence cascades. Soft Matter 16, 1866.
6. Chudak, M. K., Chopra, V., Hensel, R., Arzt, E. and Darhuber, A.A. (2020) Elastohydrodynamic dewetting of thin liquid films – elucidating underwater adhesion of topographically patterned surfaces. Submitted for publication.