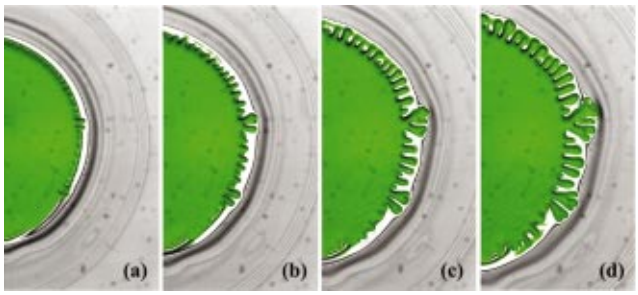

FIG. 1. A droplet of glycerol containing sodiumdodecylsulfate spreading on a glycerol film. The images were recorded at (a) $t=20$, (b) 28, (c) 36, and (d) 44 s after droplet deposition. Vertical image dimension=1 cm.

Marangoni Driven Structures in Thin Film Flows

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The spreading of a surfactant solution on a thin viscous film is dominated by Marangoni forces, which shear the liquid in proportion to the local surface concentration gradient. In conjunction with capillary forces, these tangential stresses create complex surface profiles whose fronts develop highly ramified patterns.[1,2] Theoretical models[3–6] indicate significant disturbance amplification and lateral undulations in regions of the film undergoing surfactant buildup by local pressure gradients or rapid film thinning. Figure 1 shows an example of such patterns for a droplet (green) of glycerol containing sodiumdodecylsulfate (SDS) spreading on a pure glycerol film (gray), which was spin coated onto a silicon wafer at 2000 rpm for 50 s at 46% relative humidity. The fingers elongate, widen laterally and exhibit repeated tip splitting. At the later times shown in Fig. 2, the spreading film has produced three unstable fronts (marked in green, yellow, and magenta). The magenta region corresponds to the thicker curved rim seen in Fig. 1(d) just ahead of the fingering front. The undulations (magenta) eventually separate into discrete droplets. Figure 3 shows the edge of a droplet of polydimethylsiloxane (PDMS) spreading on a film of tetra(ethylene glycol). The PDMS front steepens and develops periodic undulations that evolve into bulges whose amplitude increases with time. These fronts never develop the highly bifurcated structures shown in Fig. 2. Studies are underway to quantify the spreading patterns in terms of the liquid–liquid solubility, viscosity ratio, surface tension difference, and droplet to film aspect ratio.

The spin coated films ranged in thickness from 1–10 μm. Droplets formed at the tip of a very fine wire were made to contact the liquid film thereby initiating the spreading process. All images were obtained with an Olympus BX-60 microscope equipped with a bandpass filter centered at a wavelength of 535 nm. The light and dark bands represent contour lines of equal film thickness. Dark spots in the images were caused by small particulates on the dust cover protecting the spreading film.

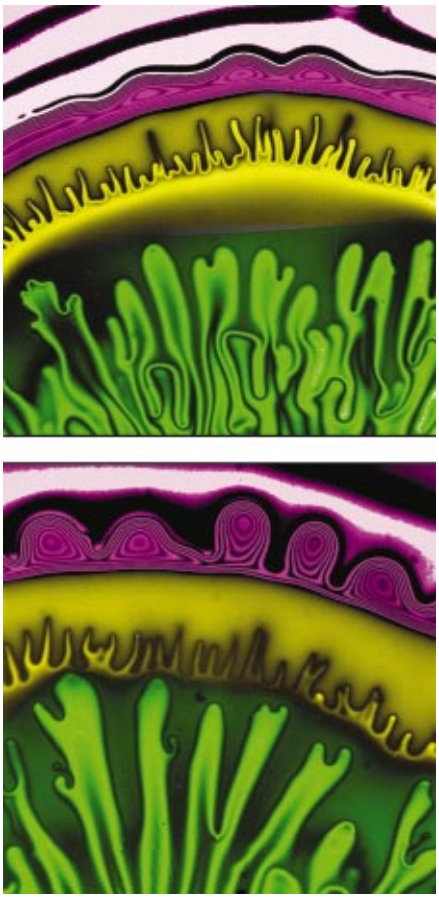

FIG. 2. A droplet of glycerol containing SDS spreading on a glycerol film. The images were recorded 5542 and 11 565 s after droplet deposition. Vertical image dimension=1 cm.

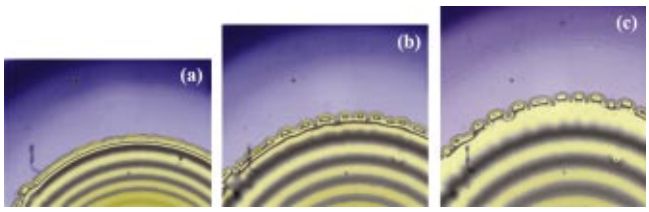

FIG. 3. A droplet of PDMS spreading on a tetra(ethylene glycol) film. The images were recorded at (a) $t=2611$, (b) 3811, and (c) 5011 s after droplet deposition. Lateral image dimension=1 cm.

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[1] S. M. Troian, X. L. Wu, and S. A. Safran, Phys. Rev. Lett. **62**, 1496 (1989).
[2] S. He and J. Ketterson, Phys. Fluids **7**, 87 (1995).
[3] S. M. Troian, E. Herbolzheimer, and S. A. Safran, Phys. Rev. Lett. **65**, 333 (1990).
[4] J. M. Davis, B. J. Fischer, and S. M. Troian, in *Interfacial Fluid Dynamics in Physicochemical Phenomena*, edited by R. Narayanan, *Lecture Notes in Physics* (Springer-Verlag, New York, 2003), and references therein.
[5] B. J. Fischer and S. M. Troian, Phys. Rev. E **67**, 016309 (2003).
[6] B. J. Fischer and S. M. Troian, Phys. Fluids (to be published).