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Soap films as two-dimensional classical fluids

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We present a theoretical derivation of the equations governing the motion of liquid soap film taking into account the physical dynamic as well as the chemical one. Generally, the behavior is specific to soap film, but we define some domains of the parameter space where its dynamic corresponds to classical two-dimensional incompressible flows. In this case we give an equation for the thickness of the membrane. We then simulate on a computer various regimes to explain how the thickness is linked to the internal motion.

Soap films and soap bubbles are fascinating objects; their properties (stability, elasticity, ...) are well known from early works by Plateau[1] and Gibbs.[2] Extensive reviews may be found in Mysels, Shinoda, and Frankel[3] and Boys[4] mainly for experiments, in Rusanov and Krotov[5] for thermodynamic theory.

Recently, a new experimental field dealing with the two-dimensional hydrodynamics of liquid soap films has been introduced by Couder and co-workers[6–12] and Gharib and Derango.[13] They observed that the thickness varies with the velocity field and therefore provides a practical, instantaneous, and global visualization of the flow structure. This new type of experimental technique raises several questions, such as the following: Is there a regime where the soap film behaves as a two-dimensional Newtonian fluid and how does the thickness depend upon the motion?

The full dynamics of soap film is in general very complex; it includes chemical kinetics between multicomponent phases out of global and local equilibrium (Fig. 1), three-dimensional motion inside the film and inside the air. In a previous paper, Couder, Chomaz, and Rabaud[8] have deduced a set of simplifications from experimental results that we are going to adopt. First, we need to describe the equilibrium state of a membrane submitted to a fixed stretch defined by a surface tension σ. The film is composed by a bulk phase and a double surface phase which may exchange soap molecules (Fig. 1). We treat the case where only one tensioactive species is present. Its surface concentration Γ_1 and the bulk concentration c_1 are linked by

$$c_0 = c_1 + 2\Gamma_1/h\,, \tag{1}$$

where c_0 is the total concentration of soap, given by the initial solution from which the membrane has been made. h represents the homogeneous thickness of the membrane. The chemical potential of the soap molecules are equal in the bulk and in the surface phase. In this simple case, this imposes a relation between Γ_1 and c_1 which will be taken as linear,

$$\Gamma_1 = K c_1\,, \tag{2}$$

where the parameter K depends only on the kind of soap you are working with, and it corresponds to an equivalent thickness of the surface. Naturally, the surface tension depends on the soap molecules adsorbed on the surface. Ideally it verifies a perfect gas law

$$\sigma = \sigma_0 - \Gamma_1 RT\,, \tag{3}$$

where R is the usual perfect gas constant and T the temperature. Equations (2) and (3), which seem quite crude at first, in fact, fall in with the experiments.

To fully understand the dynamic of soap films it is necessary to describe the response of a film to an instantaneous change of surface tension. The sequence of events is idealized in Fig. 1. When the tension applied to a film in chemical equilibrium is increased [Fig. 1(a)], it stretches, the surface density of soap decreases, and thus the surface tension increases and finally balances the extra force [Fig. 1(b)]. In the process, no molecule has enough time to migrate from the bulk to the surface. After the chemical relaxation time τ^*, the two phases go back to the chemical equilibrium [Fig. 1(c)]. The instantaneous response defines the so-called Marangoni elasticity and the long-time response the Gibbs elasticity. The Marangoni elasticity allows propagation of elastic waves (equivalent

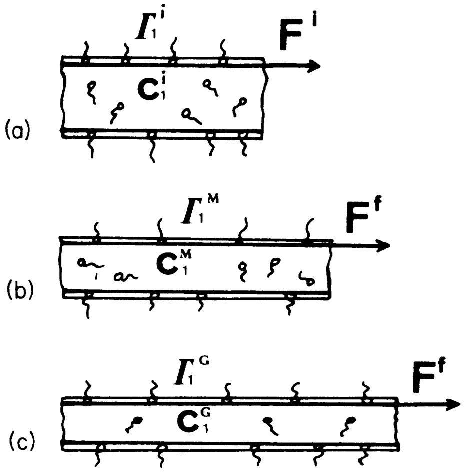

FIG. 1. Evolution of a membrane instantaneously stretched (the force applied to the side goes from F^i to F^f) with a fixed width L: (a) initial state, (b) instantaneous response (Marangoni elasticity), and (c) asymptotic response (Gibbs elasticity). The soap molecules have migrated from the bulk fluid to the surface.

of sound waves in gases) at a velocity v_L ($v_L = [2RTc_0K/\rho(H+2K)]^{1/2}$, 2ρ being the density of water, H being the mean thickness). The chemical kinetics define a time scale τ^* which depends on the particular setting. If the diffusions inside the bulk fluid were the only limiting phenomenon[5] τ^* would be roughly 10^{-2} s for a value of h of about 10^{-5} m. In fact, experimental results[8] impose 1 s at least for τ^* or more which may be due to a small contamination of the surface.[5] Thus, the chemical kinetics act on the same time scale as the dynamics and must be taken into account. For a homogeneous membrane the kinetic equation for Γ_1 will be assumed as linear. All dependencies on other parameters are pushed back in $\tilde{\Gamma}_1$, the equilibrium concentration [given by (1) and (2)],

$$d\Gamma_1/dt = -(\Gamma_1 - \tilde{\Gamma}_1)/\tau^* . \tag{4}$$

This equation specifies τ^* which characterizes the tensioactive component used. Other legitimated hypothesis (as taking Kc_1 instead of $\tilde{\Gamma}_1$) will lead to the same final first-order equation [with $2K\tau^*/(H+2K)$ instead of τ^*].

Because of the different scales of the thickness of the film (a few micrometers) and of the typical extension of the motion (a few millimeters), we may neglect any differential transport inside the film. This means that we may assume the velocity to be in the plane of the film and uniform in depth (plug flow); we may also assume that no exchange of chemical species occurs between different points of the film. The chemical equilibrium or kinetics is therefore a local motion. Given this fundamental assumption, we may now speak about particles of the film which are characterized by a well-defined mass and composition. Writing the Newton law for this particle, we get the dynamical equation. In the absence of internal forces its expression reads

$$\rho h \, d\mathbf{u}/dt = 2\nabla\sigma + h\mu\Delta\mathbf{u} , \tag{5}$$

where h is the local thickness of the film, ρ the density of the solution, μ its viscosity, and $\mathbf{u}$ the two-dimensional velocity. To simplify further, the surface layer viscosity has been neglected. As we will consider only small variation of h, its introduction would just change μ into an effective viscosity. The surface tension σ is assumed to be equal on both sides. The Lagrangian derivative is represented by d/dt. From the conservation of masses, we obtain the following:

$$dh/dt = -h\nabla\cdot\mathbf{u} , \tag{6}$$

$$dc_0/dt = 0 . \tag{7}$$

Taking into account molecules transport and flux, Eq. (4) becomes

$$d\Gamma_1/dt = -\Gamma_1\nabla\cdot\mathbf{u} - [\Gamma_1 - c_0hK/(h+2K)]/\tau^* . \tag{8}$$

The set of Eqs. (1)-(3), (7), and (8) is now closed. Although it is not our purpose here, it may simply be adapted to take into account gravity effect. We claim that this set contains all the physics of a liquid soap film. We take the case where c_0 is initially uniform and remains so because of the conservation law (7). The case of a nonuniform c_0 is important and will be treated in a forthcoming paper. Scaling (u,x,t,h,Γ_1) by typical values $[U,L,L/U,H,c_0KH/(H+2K)]$, we obtain

$$\begin{aligned} d\mathbf{u}/dt &= -M^{-2}h^{-1}\nabla\Gamma_1 + R_e^{-1}\Delta\mathbf{u} , \\ d\Gamma_1/dt &= -\Gamma_1\nabla\cdot\mathbf{u} - [\Gamma_1 - h(1+2k)/(h+2k)]/\tau , \\ dh/dt &= -h\nabla\cdot\mathbf{u} , \end{aligned} \tag{9}$$

with $k = K/H$, $R_e = UL\rho/\mu$, and $\tau = \tau^* U/L$. M is the analogous to the Mach number, U/v_L. By dimensional analysis on (9) we see that the relative variations of Γ_1 and h are of the order M^2, thus small when the velocity is small compared to v_L. This corresponds to the experimental cases.[6–13] To understand the dynamic of soap films we may use asymptotic theory and expand variables in a series of $M^2 = \epsilon$, $a = \sum_j \epsilon^j a_j$. We get at zero order, $\Gamma_{10} = h_0 = 1$, and at first order,

$$\begin{aligned} \partial\mathbf{u}_0/\partial t + \mathbf{u}_0\cdot\nabla\mathbf{u}_0 &= -\nabla\Gamma_{11} + R_e^{-1}\Delta\mathbf{u}_0 , \\ \nabla\cdot\mathbf{u}_0 &= 0 , \end{aligned} \tag{10}$$

which is the Navier-Stokes system for an incompressible fluid. We deduce that Γ_{11} represents a pressure. The next order gives the following equations for h_1:

$$h_1 = \Gamma_{11} + e/2k , \tag{11}$$

$$\partial e/\partial t + \mathbf{u}_0\cdot\nabla e = -(\Gamma_{11} - e)\tau'^{-1} . \tag{12}$$

The variation of the thickness splits in two parts: The pressure part Γ_{11} of (11) corresponds to an instantaneous Marangoni stretching and the $e/2k$ contribution corresponds to the relaxation toward the Gibbs equilibrium. For a specified flow the field e depends only on τ', $\tau' = \tau(1+2k)/2k$, a measure of the local chemical relaxation time scaled by the advection time. For a fast relaxation $\tau' \ll 1$, e is equal to the pressure and h_1 follows the Gibbs relation $h_1 = \Gamma_{11}(2k-1)/2k$.

In the thickness expression (11), e is weighted by $1/2k$ which compares the amount of soap molecules in the bulk and the one adsorbed on the surfaces. Pure Marangoni dynamics ($h_1 = \Gamma_{11}$) are obtained when the surfaces are predominant ($k \gg 1$).

In those two limits, pure Gibbs or pure Marangoni dynamics, the thickness just visualizes the pressure field. Figure 2 shows at time $tU/L = 16$, the vorticity and pressure fields of a vortex couple generated by the secondary instability of a Bickley jet at an actual Reynolds number

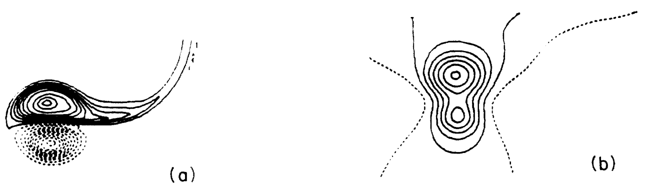

FIG. 2. Vortex couple produced after $T = 16$ by a numerical simulation of the Bickley jet for $\mathcal{R} = 300$ with a spectral code 128^2 ($L = 2\pi$, $U = 1$, initial wake thickness $L_j = 0.3$): (a) the vorticity, isolines every 0.4 by 0.4; (b) the pressure equivalent Γ_{11}, isolines every 0.025 by 0.025.

($\mathcal{R} = R_e L_j$, L_j being the width of the jet) of 300. We use a spectral numerical code 128^2 (see Ref. 7 for details). As the pressure is a nonlocal quantity it "feels" globally the flow and smoothes out all the details [Fig. 2(b)]. Such a field [Fig. 2(b)] does not correspond to observations made with soap films.[9] We need the memory effect given by the chemical relaxation. We compute for this prototype simulation the field e using Eq. (11). Figure 3 presents the fields e for various relaxation time τ'. For $\tau' \ll 1$ the pressure field is recovered. But as τ' increases and becomes of order 1, e starts to "look like" the modulus of the vorticity. The two vortices that form the couple, while in the same depression, form two separate structures in Figs. 3(b) and 3(c). Going back to the thickness, we deduce from Eq. (11) that the correlation between h_1 and the vorticity field, when $\tau' \gtrsim 1$, will be stronger if k is small (e.g., an equilibrium dominated by the bulk phase).

In conclusion, we may say that soap films will behave like a two-dimensional incompressible fluid as soon as we run experiments at a low speed U compared to the velocity ($v_L \sim 5$ m/s) of longitudinal Marangoni waves (equivalent of sound waves in gases). In this case, the relative variations of thickness are small and of order $(U/v_L)^2$, the equivalent Mach number. The thickness field will follow the two-dimensional pressure field in the limits of extremely clean solution (rapid equilibrium thus Gibbs dynamics) or of a thin film where there is negligible relaxation (Gibbs and Marangoni dynamics equivalent). In the other cases the thickness will be linked by an noninvertible relation to the velocity field, but will provide a better visualization of motion for slow chemical relaxation ($\tau' \gtrsim 1$) and thick film (small k).

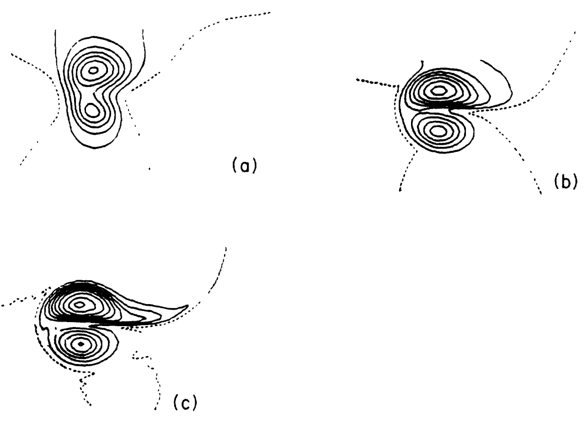

FIG. 3. The e component of the soap film thickness (see Fig. 2) vs τ' (to be compared with 3, the order of U/L_j): (a) $\tau' = 0.2$, isolines every 0.02 by 0.02; (b) $\tau' = 2$, isolines every 0.025 by 0.025; (c) $\tau' = 10$, isolines every 0.025 by 0.025.

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[1] J. Plateau, *Statique Expérimentale et Théorique des Liquides Soumis aux Seules Forces Moléculaires* (Gauthier Villars, Paris, 1873).

[2] J. W. Gibbs, *The Collected Works* (Longmans Green, New York, 1931).

[3] K. J. Mysels, K. Shinoda, and S. Frankel, *Soap Films Studies of Their Thinning* (Pergamon, New York, 1959).

[4] C. V. Boys, in *Soap Bubbles and the Forces which Mould Them* (Society for Promoting Christian Knowledge, London 1890; Anchor, New York, 1959).

[5] A. L. Rusanov and V. V. Krotov, Prog. Surf. Membr. Sci. **13**, 415 (1979).

[6] C. Basdevant, Y. Couder, and R. Sadourny, in *Macroscopic Modelling of Turbulent Flows*, edited by U. Frisch *et al.*, Lectures Notes in Physics, Vol. 230 (Springer-Verlag, Berlin, 1984).

[7] J. M. Chomaz, M. Rabaud, C. Basdevant, and Y. Couder, J. Fluid Mech. **187**, 115 (1988).

[8] Y. Couder, J. M. Chomaz, and M. Rabaud, Physica D **37**, 384 (1989).

[9] Y. Couder and C. Basdevant, J. Fluid Mech. **173**, 225 (1986).

[10] Y. Couder, C. Basdevant, and H. Thome, C. R. Acad. Sci. **299**, 89 (1984).

[11] Y. Couder, J. Phys. (Paris) Lett. **45**, 353 (1984).

[12] Y. Couder, J. Phys. (Paris) Lett. **42**, 429 (1981).

[13] M. Gharib and P. Derango, Physica D **37**, 406 (1989).