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"Marginal pinching" in soap films

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Abstract. – We discuss the behaviour of a thin soap film facing a frame element: the pressure in the Plateau border around the frame is lower than the film pressure, and the film thins out over a certain distance $\lambda(t)$, due to the formation of a well-localized pinched region of thickness $h(t)$ and extension $w(t)$. We construct a hydrodynamic theory for this thinning process, assuming a constant surface tension: Marangoni effects are probably important only at late stages, where instabilities set in. We find $\lambda(t) \sim t^{1/4}$, and for the pinch dimensions, $h(t) \sim t^{-1/2}$ and $w(t) \sim t^{-1/4}$. These results may play a useful role for the discussion of later instabilities leading to a global film thinning and drainage, as first discussed by K. Mysels under the name "marginal regeneration".

Early experiments by K. Mysels and coworkers [1] showed that a vertical soap film, suspended on a frame (and made with "mobile" surfactant), thins out by nucleation and growth of black, thin spots near the Plateau borders. They called this process "marginal regeneration".

There are in fact (at least) two steps in marginal regeneration: a) The pressure in the Plateau border is lower than the pressure in the film. This thins out the film near the border, and leads to a "pinch". b) The pinched state must have an intrinsic instability leading to the black spots. The two steps are very different: the pinch can occur at constant surface tension —*i.e.* without any Marangoni effect. On the other hand, the later instabilities are probably triggered by Marangoni flows, as pointed out by a number of authors [2–5].

Our aim in the present note is restricted to the first step, *i.e.* the description of the pinched state with its dynamics. Pinching has already been studied in connection with the elimination of dimples in the coalescence of drops [6,7] or in the drainage of thin films [8,9]. However, in these problems, the extension of the dimpled zone is prescribed and fixes one of the spatial scales involved. Our case is different: we consider a semi-infinite film (of initial thickness e_0) facing a straight Plateau border. At $t = 0$ the film begins to pinch, and is perturbed over a certain distance $\lambda(t)$ increasing with time (as we shall see, $\lambda(t) \sim t^{1/4}$). Also the width $w(t)$ of the small pinched region decreases with time ($w(t) \sim t^{-1/4}$) and so does the thickness of the pinch $h(t) \sim t^{-1/2}$. It seems important to know these scales in detail before embarking

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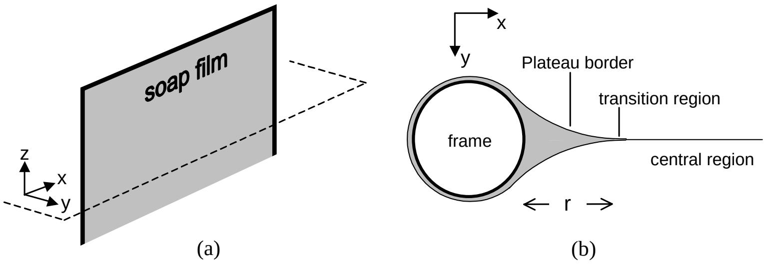

Fig. 1 – (a) Soap film held in a vertical frame, and xy cross-section plane. (b) Sketch of the initial shape in the cross-section plane (here, a case where the liquid totally wets the frame material). As the film in the central region is very thin (uniform half-thickness $e_0 \ll r$), it has been represented simply as a line.

into the second step: all data on dimples suggest that instabilities can only occur after a long stage of pinching. This is why we study the pinch in the simplest condition of a constant surface tension. In this case, as will be shown in this article, a complete, explicit solution involving the two scales λ and w can be constructed.

We thus consider a soap film, held in a vertical rectangular frame as in fig. 1a, and we wish to describe the time evolution of the film profile in a horizontal cross-section, neglecting in the process all drainage in the vertical direction (vertical drainage in "mobile" films is tightly associated with the second step of marginal regeneration, and will intervene significantly only once the instabilities have risen up). The cross-section profile is therefore treated as a one-dimensional thin film of half-thickness $e(x,t)$ at position x and time t.

The initial shape of the film has the following general features [1] (see fig. 1b): joining the frame legs is the "Plateau border" of size r, where the thickness is of macroscopic dimensions. In this region, the curvature of the film is prescribed to a fixed value c. Further away from the frame legs, there is a transition region, where the thickness decreases steeply. And, finally, we reach the zero-curvature central region, of constant thickness $2e_0$, which extends over the major part of the frame width. The system displays in addition a natural separation of spatial scales. Firstly, the width of the frame (*e.g.*, 10 cm) is supposed to be much larger than the Plateau border extension r (*e.g.*, 1 mm). We shall thus assume that the width of the frame can safely be taken as infinite relative to the length r, leaving us with a semi-infinite film. Secondly, the Plateau extension r itself is much larger than the limit thickness e_0 in the central region (*e.g.*, 10 μm). Our study of the film dynamics will therefore rely on an asymptotic approach ([1]) valid in the limit $\varepsilon \ll 1$, where ε represents the aspect ratio of the film:

$$\varepsilon = e_0/r\,. \tag{1}$$

The evolution equations are classically written in the lubrication approximation. The axes are shown in fig. 1b, and, for simplicity, we choose the origin of the x-axis at the position of minimum thickness, in the pinch ([2]). Assuming that the velocity of the liquid vanishes at the film/surfactant layer interface, the flow is of Poiseuille type and yields a parabolic velocity profile. With a Laplace-Young pressure $p = p_{\text{air}} - \gamma e_{xx}$ in the fluid, we find that the flux Q

([1])This kind of approach has been initiated and developed by Jones and Wilson [7] in the resolution of a closely related coalescence problem.

([2])We assume that, once the pinch is formed, its spatial position does not change anymore, or, at least, that its motion remains of subdominant importance to the solution. This hypothesis is indeed supported by previous work on dimple formation in analogous situations [6, 8, 9].

in the x-direction is given by $Q = \frac{2}{3}V^* e^3 e_{xxx}$. Here, $V^* = \gamma/\eta$ is a characteristic capillary velocity and is defined as the ratio of the surface tension γ by the viscosity of the liquid η. (γ might depend on the vertical elevation of the considered cross-section, but is otherwise constant in the xy-plane.) Inserting it into the continuity relation $2e_t + Q_x = 0$, we find the governing equation of the film profile:

$$e_t + \frac{V^*}{3}(e^3 e_{xxx})_x = 0\,. \tag{2}$$

Note that microscopic interactions [10] like van der Waals, or electrical interactions, have not been included in eq. (2), since we are mainly interested in thicknesses larger than their range of action. We will simply consider that once the film thickness reaches small enough values ($\simeq 100\,\text{Å}$), these additional interactions are globally repulsive and prevent any further thinning.

It is more convenient to use the reduced variables (denoted with capital letters) $X = x/r$, $E = e/e_0$, and $T = t/t_{\text{relax}}$, where $t_{\text{relax}} = \frac{3}{V^*} r^4/{e_0}^3$ is a typical relaxation time needed for a bump of height e_0 and width r to be leveled off by capillary flow ([3]). In these reduced variables, eq. (2) takes the dimensionless form

$$E_T + (E^3 E_{XXX})_X = 0\,. \tag{3}$$

From the initial shape described above, a pinch has first to form. Describing this process requires the precise knowledge of the initial state. However, it is expected that the formation stage is quite rapid on the time scale of t_{relax}, and, moreover, the details of it appear in fact to be unessential to the longer-term dynamics of the film. We may nevertheless understand why only a localized region thins out at the beginning and starts to pinch: within the initial profile, the pressure gradient is non-vanishing only over a very restricted zone, inside the transition region (there, the curvature decreases from the border value c to the central value zero).

We will hereafter simply assume that this early stage leads to the formation of a pinch precursor of reduced thickness. Without the need of further knowledge, we are then in a position to describe the evolution of the soap film. In order to do so, we solve the equation governing the dynamics (eq. (3)) as follows: we divide the film into three spatial regions (inherited from the three regions in the initial profile), and look for solutions in each of them, imposing in addition that these solutions all match together (in the sense of asymptotic matching). Starting from the frame leg, the three regions are: the Plateau border (at negative values of X), then the pinch region that lies in the vicinity of $X = 0$, and, eventually, the central region, which extends over the positive values of X.

The profile in the Plateau border is easily determined to leading order in the asymptotic parameter ε: as the border contains a great mass of liquid compared to the rest of the film, it remains unchanged during the whole evolution. In particular, its curvature remains the same as initially (*i.e.*, $e_{xx} = c$), maintaining thereby a constant pressure drop as compared to the pressure in the central region. This feature will naturally prove essential to the film dynamics.

For clarity's sake, it turns out to be more convenient to pursue the description with the central region ($X > 0$). The existence of a pinch at $X = 0$, with a sudden thickness decrease, involves that the central region, initially flat, is pushed out of equilibrium, and that a flow must take place in order to relax, to the greatest possible extent, the curvature induced by such pinching. But this flow cannot easily pour liquid through the pinch region, whose small

([3])Here is a simple way to see this: the capillary flux, as used in establishing eq. (2), is $Q \simeq V^* e^3 e_{xxx} \sim V^* {e_0}^4/r^3$. Then, t_{relax} appears as the time needed to empty the volume of the bump with such a flux, *i.e.*, $t_{\text{relax}} \simeq e_0 r/Q$.

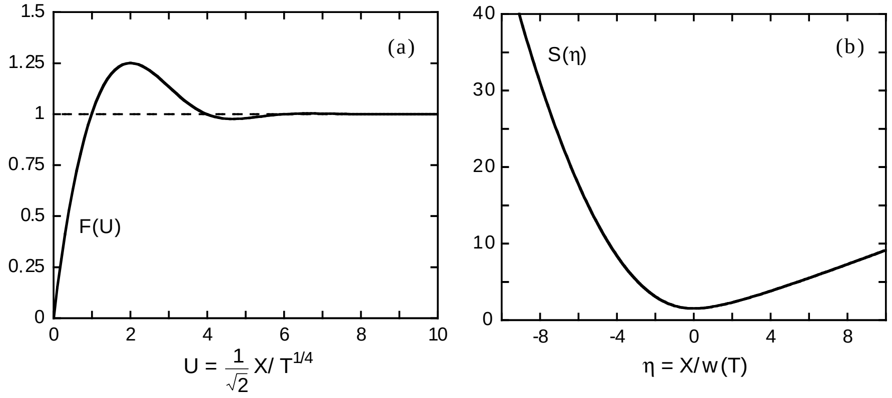

Fig. 2 – (a) The self-similar function $F(U)$ describing the film profile in the central region. (b) The self-similar function $S(\eta)$ describing the pinch.

dimensions strongly reinforce viscous resistance. Thus, the relaxation flow is mostly directed towards the far-field zone "at infinity" ($X \gg 1$). In a more formal way, the relaxation flow Q_{relax} towards the far-field zone is of order unity, whereas the flow poured through the pinch Q_{pinch} is only of order ε (as will be shown later on). Thus, in the spirit of boundary-layer techniques, we look inside the central region for a solution at zeroth order in ε (a so-called "outer" solution), that is to say that the relaxation process is solved with a boundary condition of *zero* flux at $X \ll 1$ (in the vicinity of the pinch region). The complete set of boundary conditions for the central region solution writes:

$$\text{i)} \quad E|_{X\ll 1} = 0\,, \qquad \text{ii)} \quad E^3 E_{XXX}|_{X\ll 1} = 0\,, \qquad \text{iii)} \quad E|_{X\gg 1} \to 1\,. \tag{4}$$

The first condition imposes that the film thickness must vanish (at zeroth order in ε) at the approach of the pinch, the second one is the zero flux condition, and the third one ensures that the far-field film remains unperturbed and retains the initial thickness e_0.

We propose a solution to eqs. (3) and (4) in a self-similar form, with the self-similar variable $U = \frac{1}{\sqrt{2}} X/T^{1/4}$. Setting $E(X,T) = F(U)$, and inserting it into eq. (3), we obtain the following equation for the self-similar function F:

$$UF' = (F^3 F''')'\,. \tag{5}$$

A study of the film profile in the pinch region (see further down) shows that a matching with the central region is possible if the behaviour of F near $U = 0$ is either linear, parabolic, or power law (with exponent 3/4). However, it turns out that only the linear behaviour is compatible with eqs. (4) and (5), *i.e.*, $F \sim AU$ for $U \ll 1$, with A a constant. The complete function F can be constructed numerically with the help of a two-sided shooting procedure based on analytical expansions at $U \ll 1$ and $U \gg 1$ (we omit technical details for conciseness). The resulting plot is given in fig. 2a: as one can see, F increases from zero to unity over a range of a few units in the variable U, and then presents decreasing capillary oscillations. The shooting procedure also determines the numerical value of the slope of F at the origin: $A \simeq 1.591$.

The important physical consequence of these results is that, because of the presence of the pinch at $X = 0$, the film in the central region is also thinned out, and retrieves the far-field thickness e_0 over a distance $\lambda(t) \simeq rT^{1/4} \sim t^{1/4}$, which increases with time.

We are thus in possession of a self-similar solution of the problem in the central region. One may though wonder about the relevance of self-similarity in describing the real physical solution. Actually, if we take a version of eq. (3) that is linearized around $E = 1$, and solve it analytically with the Green function method with boundary conditions similar to eq. (4), we rigorously find that the solution indeed takes the above self-similar form. We conjecture here that this feature can be safely carried over to the non-linear case.

We next consider the solution in the pinch region. As stated earlier, once the pinch is well-formed, the suction from the Plateau border is able to drain liquid through the pinch out of the central region, but only with an asymptotically small current Q_{pinch}. However, the dimensions of the pinch are so small that we can reasonably take this current to be spatially uniform over it [6, 7, 9] (but not constant in time): $Q_{\text{pinch}} = Q_{\text{pinch}}(T)$. Therefore, to leading order, the profile of the film in the pinch region obeys the following uniform current equation, rather than the full eq. (3):

$$E^3 E_{XXX} = Q_{\text{pinch}}(T)\,. \tag{6}$$

Here again, we try a self-similar solution of the form $E(X,T) = h(T)S(\eta)$, with $\eta = X/w(T)$. Such a solution has been proposed in refs. [7,9]. In this solution, the functions $h(T)$ and $w(T)$ represent, respectively, the typical *height* and *width* of the pinch, and will be determined by matching with the neighbouring Plateau and central regions. Replacing the self-similar form in eq. (6), we get $S^3 S'''\, h(T)^4/w(T)^3 = Q_{\text{pinch}}(T)$, from which we deduce that S^3S''' cannot bear any dependence on η (this would bring in the l.h.s. of the equation a dependence on X that could not be balanced by the r.h.s.). Thus, the differential equation on S is necessarily

$$S^3 S''' = -\alpha\,, \tag{7}$$

where α is a positive constant (the sign has been chosen in view of the fact that Q_{pinch}, oriented towards the negative coordinates, is negative). At this point, α is still unknown, but shall be determined later on.

Let us now apply the matching requirements that the self-similar solution must fulfill, starting by the match with the Plateau border. A study of the asymptotic behaviours permitted to S by eq. (7) for large η (either positive or negative) opens three possibilities, namely linear, parabolic or power law with exponent 3/4. The Plateau border imposes a constant curvature $e_{xx} = c$, or in terms of the reduced variables, $E_{XX}|^{\text{Plateau}}_{X\to 0} = rc/\varepsilon$. Thus, on the pinch side, S must be asymptotically parabolic. Equating the Plateau curvature with the pinch curvature $E_{XX}|^{\text{pinch}}_{\eta\to-\infty} = \lim S''\, h(T)/w(T)^2$ yields moreover the two conditions

$$\text{i)}\quad h(T)/w(T)^2 = rc/\varepsilon\,, \qquad \text{ii)}\quad \lim_{\eta\to-\infty} S''(\eta) = 1\,. \tag{8}$$

We proceed further and match now on the other side, with the central region. As stated above, there are three possibilities of asymptotic behaviour of S at the approach of the central region, but inside the central region we found that only the linear one is consistent for the function F with the boundary conditions (4) and the differential equation (5), giving $E(X,T)|^{\text{central}}_{X\ll 1} \simeq AU = \frac{A}{\sqrt{2}} X/T^{1/4}$. In the pinch, $S(\eta)$ must accordingly have an asymptotically linear behaviour (for $\eta \gg 1$), which leads to the corresponding expression of E: $E(X,T)|^{\text{pinch}}_{\eta\gg 1} = h(T)\,\eta\,\lim S' = X\,h(T)/w(T)\,\lim S'$. Matching these central and pinch behaviours together, we find that two new conditions must be met:

$$\text{i)}\quad h(T)/w(T) = \frac{A}{\sqrt{2}} T^{-1/4}\,, \qquad \text{ii)}\quad \lim_{\eta\to+\infty} S'(\eta) = 1\,. \tag{9}$$

With eqs. (8-i) and (9-i), we are now able to extract the expressions of h and w:

$$h(T) = \varepsilon\,\frac{A^2}{2rc}T^{-1/2}\,, \qquad w(T) = \varepsilon\,\frac{A}{\sqrt{2}rc}T^{-1/4}\,. \tag{10}$$

We immediately notice that, as announced before, h and w, the typical dimensions of the pinch, are of order ε and hence asymptotically smaller than the dimensions (of order unity) that prevail in the central region. Furthermore, we see that, as time passes, the pinch becomes thinner and smaller in extent. We may also note that the time dependences in eq. (10) are, not too surprisingly, equal to the ones found in closely related drainage problems [6, 7, 9].

To complete our solution of the profile in the pinch, we still have to exhibit a function S that has the correct shape, as dictated by eqs. (7), (8-ii) and (9-ii). Such a function has indeed been computed previously [6,7]. At large η, S admits an expansion around the imposed linear profile of slope unity, from which we have started numerical integrations towards negative values of η. The integrations have shown that complying with the requirement that S'' equals unity for large negative η ascribes to the parameter α in eq. (7) the value $\alpha \simeq 1.21$. The minimum value of S (at $\eta = 0$) is then found to be $S_{\min} \simeq 1.52$. A plot of S is provided in fig. 2b.

With the knowledge of the self-similar functions F and S, we have now at disposal a complete description of the profile of the soap film, once the pinch is formed (and under the basic hypothesis that it remains later on fixed in position). We may at this point give justifications to some assumptions that were made in the course of the resolution.

First, it was assumed that to zeroth order in ε, one was entitled to solve the central region profile with zero flux towards the pinch (eq. (4-ii)). This can be justified as follows. In the central region, the current generated by the relaxation process is, by the very definition of the reduced variables, of order one: $Q_{\text{relax}} = \mathcal{O}(1)$. On the other hand, the expression of the current in the pinch was given when establishing eq. (7): $Q_{\text{pinch}} = S^3 S'''\, h(T)^4/w(T)^3$. Since both h and w are of order ε, we deduce that $Q_{\text{pinch}} = \mathcal{O}(\varepsilon) \ll Q_{\text{relax}}$, and is thus negligible to leading order in the central region.

A second approximation was that, since the pinch was of small dimensions, the current Q_{pinch} could be taken uniform inside it. Indeed, using the continuity equation, one can estimate the thereby neglected spatial variation δQ_{pinch} as the variation in time of the volume enclosed in the pinch region: we have $\delta Q_{\text{pinch}} \simeq \frac{\mathrm{d}}{\mathrm{d}t}(wh) = \mathcal{O}(\varepsilon^2) \ll Q_{\text{pinch}}$, as had been assumed.

In addition, it has been assumed, when establishing eq. (2), that the velocity field goes to zero at the film/surfactant layer interface, but also that the surface tension γ keeps a constant value throughout the film. This cannot be strictly true: For the surfactant layer to be at rest, it must develop a surface tension gradient to counter the friction force generated by the underneath liquid flow. We can estimate the required gradient by equating it with the viscous stress at the surface: $(1/\gamma_0)\partial\gamma/\partial x = (\eta/\gamma_0)\partial v/\partial y|_{y=e} = e e_{xxx}$ (the equations were divided through by γ_0, the constant tension that would be obtained in the absence of liquid flow). Using this formula with our solution for $e(x,t)$, we find that the relative variation of the surface tension across the pinch is $\delta\gamma/\gamma_0|_{\text{pinch}} = \int_{\text{pinch}} (1/\gamma_0)\partial\gamma/\partial x\, \mathrm{d}x \simeq w({e_0}^2/r^2)(h^2/w^3) = \mathcal{O}(\varepsilon^2)$. Similarly, we find that the variation over the characteristic length r in the central region writes $\delta\gamma/\gamma_0|_{\text{central}} = \mathcal{O}(\varepsilon^2)$. Thus, the gradients required to cancel the flow velocity are indeed very small, and the surface tension can safely be taken as constant (at least, in the study of the leading-order dynamics of the film). However, we should point out that this result is by no means contradictory with the later destabilization by Marangoni flows, as these occur in the *vertical* direction.

A final remark is in order concerning the late stage of the film pinching. As mentioned earlier, we expect that, at small scales, repulsive microscopic forces stop the thinning (with different possible scenarii [8] depending on the electrolyte concentration in the film). The corresponding time $t_{\rm stop}$ can be roughly estimated, if we say that the stop occurs for e around 100 Å, and use eq. (10) with typical numerical values (for instance, $e_0 \simeq 10\,\mu$m, $r \simeq 1/c \simeq 1\,$mm, $V^* \simeq 10^2\,$m/s): we find $t_{\rm stop} \simeq 10^3\,$s. We should caution that, with respect to experiments on marginal regeneration, this is a significant duration, for which one would then probably have to include the study of the second step of marginal regeneration (Marangoni instabilities). The thinning process might therefore not be allowed to proceed by itself as far as $t_{\rm stop}$. We can also estimate the distance over which the central region has a thickness departing from e_0 (assuming that the thinning keeps on until $t_{\rm stop}$): $\lambda_{\rm stop} \simeq r(t_{\rm stop}/t_{\rm relax})^{1/4} \simeq 3r \simeq 3\,$mm. This distance is much smaller than the frame width, justifying our description of the film as semi-infinite towards the center of the frame. However, in situations where the frame width is not so large, our central region solution should be regarded as transient: when $\lambda(t)$ becomes of the order of the frame width, we expect to retrieve the solutions in bounded geometries that have been described by several authors [6, 7, 9] (for instance, a quasi-static parabola in the central region).

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