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Upward creep of a wetting fluid : a scaling analysis

J. F. Joanny and P.-G. de Gennes

Laboratoire de Physique de la Matière Condensée, Collège de France, 75231 Paris Cedex 05, France

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Résumé. — Un liquide de Van der Waals, montant depuis un réservoir le long d'une paroi verticale, doit montrer toute une série d'étapes : i) Un ménisque macroscopique se met en place à une échelle de temps t_1 (s) et atteint l'altitude classique $h_c \sim 1$ mm. ii) Un film précurseur dynamique apparaît à $t_2 \sim 10^{-2}$ s. L'extrémité de la « langue » avance vite jusqu'à un temps $t_3 \sim 1$ s puis avance plus lentement. iii) Aux temps $t > t_4$ ($t_4 \sim 10$ s) apparaît une portion du film statique final, entre l'altitude h_c et une altitude limite $h_l(t)$. iv) Après un intervalle $t_5 \sim 10$ jours, on arrive à un régime asymptotique, self-similaire, de croissance.

Abstract. — A Van der Waals fluid, climbing from a reservoir along a vertical wall, should display a remarkable cascade of events : i) A macroscopic meniscus is set up in a short time t_1 and reaches the classical altitude $h_c \sim 1$ mm. ii) A dynamic precursor film appears at a time $t_2 \sim 10^{-2}$ s. The tip of the « tongue » advances fast up to a time $t_3 \sim 1$ s and then more slowly. iii) A portion of static film builds up (from h_c up to a certain altitude $h_l(t)$) at times $t > t_4$ ($t_4 \sim 10$ s). iv) A self-similar, slow, growth of both the « tongue » and the static portion is reached after a time $t = t_5 \sim 10$ days.

1. General aims.

1.1 A REMINDER ON STATIC FILMS. — A wetting fluid tends to climb along a vertical wall, and to build up a film. This film is particularly conspicuous with superfluid helium (the Rollin film [1]) but is present for many fluids [2]. The equilibrium film thickness $e_0(h)$, as a function of altitude h , results from a balance between the disjoining pressure $\Pi(e)$ (describing the long range forces from the solid [3]) and the gravitational pressure [4] :

$$\Pi(e_0) = \rho gh \quad (1)$$

ρ being the density of the liquid (the density of the ambient gas phase is assumed to be negligible). Ultimately at very high altitudes, the film is truncated : it becomes more favourable to lose in wetting energy and to avoid exceedingly high gravitational energies. This is typical of a prewetting transition [5], and occurs at an altitude h_m such that [6]

$$\left[e_0 \Pi(e_0) + \int_{e_0}^{\infty} \Pi(e) de \right]_{h=h_m} = S \quad (2)$$

where S is the spreading coefficient

$$S = \gamma_{so} - \gamma_{sL} - \gamma. \quad (3)$$

Here the γ_{ij} 's are interfacial tensions between solid (s) liquid (L) and gas (o) ; we use the shorthand $\gamma_{Lo} = \gamma$ for the surface tension of the liquid.

The thickness $e_0(h_m)$ defined by equation (2) is controlled by the spreading parameter S , and will be called e_s . To make the discussion more concrete, let us consider the simplest case of *Van der Waals fluids*, attracted towards the wall by an unretarded interaction [7]. Then the disjoining pressure is

$$\Pi(e) = \frac{A}{6 \pi e^3} = \frac{\gamma a^2}{e^3} \quad (4)$$

where A is called the Hamaker constant, and has the dimensions of energy. The length $a = (A/6 \pi \gamma)^{1/2}$ is a molecular size. The static profile corresponding to equations (1) and (4) is well known [8]

$$e_0(h) = \left(\frac{a^2}{\kappa^2 h} \right)^{1/3} \quad (5)$$

where κ^{-1} is the capillary length, defined *via* $\kappa^2 = \rho g / \gamma$, and is of order 1 mm. The essential feature of (5) is a slow ($h^{-1/3}$) decrease of thickness with altitude. This is ultimately cut off at very high h ($h = h_m$) by the dewetting condition (2). But h_m is very high (for $S/\gamma = 10^{-2}$, $h_m = 10$ meters).

Two dimensionless parameters show up in these discussions

$$\kappa a = \varepsilon$$

$$\frac{S}{\gamma} = \phi^2.$$

We shall choose as typical values $a = 1 \text{ \AA}$, $\kappa^{-1} = 1 \text{ mm}$ giving $\varepsilon = 10^{-7}$, and $S/\gamma = 10^{-2}$, giving $\phi = 0.1$. The existence of these small parameters will simplify the discussion considerably. It will also turn out in section 3 that the most interesting regimes require a further inequality, namely

$$\varepsilon < \phi^3.$$

This is indeed well satisfied in all practical cases.

1.2 THE CREEP PROBLEM. — Our aim, in the present paper, is to investigate the *formation* of a film, when a vertical wall is put into contact with a bath of liquid (Fig. 1). Questions of this type occur in the physics of soils, which are partially moist and experience various thermal cycles (days, seasons). Of course, the soil problem is enormously more complex :

a) the water channels are geometrically irregular, and the films are coupled to certain macroscopic pockets of liquid (pendular droplets between two grains,...). Some consequences of this coupling have been discussed recently by one of us [9];

b) water itself is a complex fluid, for which the disjoining pressure is poorly understood [10]. In what follows we shall concentrate on the simpler case of Van der Waals fluids;

c) the *wettability* of the soil (the S parameter) has large fluctuations, which we ignore here, but which can lead to strange effects [11].

In a previous note [12] one of us discussed a related (but simpler) problem, where the solid plate is dipped, at a constant speed U , downwards into the liquid. The main emphasis was set on two limiting cases : *fast dipping* where we have no film left, and *very slow dipping* where an interesting crossover existed between a static film (at altitudes $h < h_1(U)$) and a thinner, dynamic film (at $h > h_1$). The intermediate regimes were not discussed. It turns out that for the present problem of transient climbing, the extreme regimes have their exact counterpart. However, most of the interest is in the intermediate regimes (typically for times t between 10 seconds and 10 days). We cover the fast regimes in section 2, and the longer time scales in sections 3, 4. Because of the complexity of the time sequence, our discussion will remain mostly qualitative : we construct scaling laws for the various observables, but ignore all coefficients.

Our discussion is restricted to situations where all *inertial effects are negligible* : this is correct for the processes involved, except possibly for low viscosity fluids at very early times.

2. Macroscopic contact line and contact angle.

Let us assume that we start with a strictly horizontal surface. First a macroscopic meniscus builds up (Fig. 1a). The size of the perturbed region (both horizontally and vertically) is comparable to the capillary length κ^{-1} ($\sim 1 \text{ mm}$). The ultimate height h_c of the macroscopic contact line is derived from classical capillary equilibria [13] $h_c = \kappa^{-1} \sqrt{2}$. Consider now a situation where we have not quite reached this altitude : the macroscopic contact line is at a level h_t slightly below h_c . Most of the meniscus is now at rest, and the shape can be derived from static considerations.

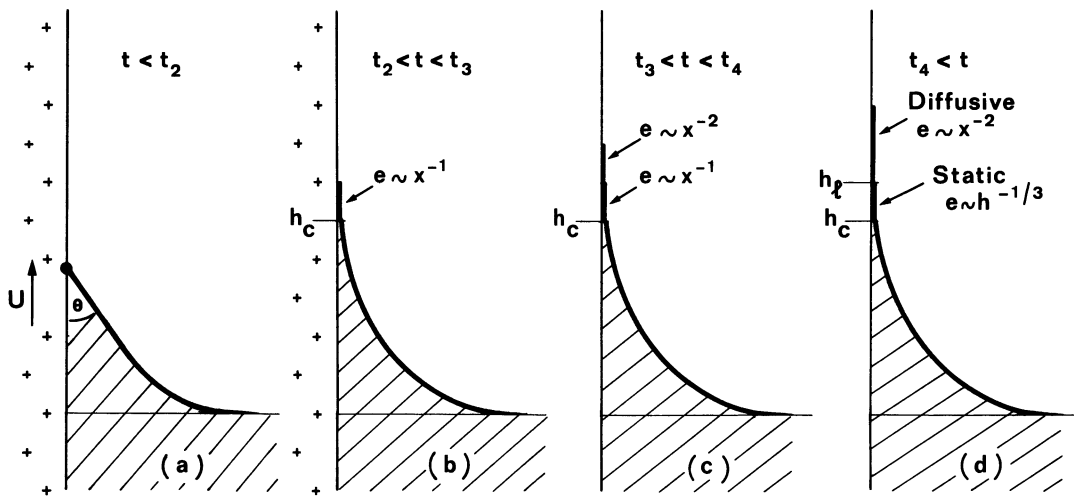


Fig. 1. — The sequence of events in vertical creep of a wetting fluid. At time 0 the fluid is assumed to be horizontal. (a) Up to a time t_2 a macroscopic profile sets up, but no precursor is present. (b) An « adiabatic » precursor appears. (c) The precursor has two parts — « adiabatic » and « diffusive ». (d) A portion of the final static film is realized. $x = h - h_c$ is the distance measured from the macroscopic limit.

In particular the (slowly varying) contact angle θ is given by the condition

$$\kappa^2 h_r^2(t) = 2[1 - \sin \theta(t)] \cong 2 - 2\theta(t). \quad (6)$$

Equation (6) can be derived from a balance of horizontal forces, as explained in [13]. We may then find the scaling law for $\theta(t)$ using the Hoffmann-Tanner relation between θ and the velocity of the contact line [14-16]

$$U = \frac{dh_r}{dt} \cong V^* \theta^3 \quad (7)$$

where $V^* = \gamma/\eta$ (η being the fluid viscosity).

Equations (6) and (7) lead to

$$\theta \cong \frac{1}{\tau^{1/2}} \quad (\tau \gg 1) \quad (8)$$

where $\tau \equiv \kappa V^* t$ is a reduced time.

Equation (8) shows that there is a long « tail » in the plot $\theta(t)$. The next question is to find when θ reaches its ultimate value which is related to the final film profile. The structure of the static film near $h = h_c$ has been studied by various authors [17-20]. The slope is of order

$$\theta \cong \frac{e_0(h_c)}{h_c} \cong \varepsilon^{2/3}. \quad (9)$$

Equating (9) and (8) we obtain an estimate of the time t_s at which the contact angle (measured at $h = h_c$) reaches its final value. In dimensionless units

$$\tau_s = \varepsilon^{-4/3}. \quad (10)$$

This corresponds to times t_s of order 10 days. We shall see later that the properties of the films change qualitatively at times shorter than t_s . But we do not expect these changes to react significantly on the contact angle law (Eq. (8)) : the macroscopic dynamics is essentially decoupled from the film dynamics [16].

A remark on inertial effects : The characteristic time displayed in equation (8) is

$$t_1 = \eta/\gamma\kappa.$$

For high viscosity fluids, this is indeed the rise

time of the meniscus, resulting from a balance between capillarity and viscosity. For low viscosities η , inertial effects come into play⁽¹⁾. The inertial rise time \tilde{t}_1 may be divided from the frequency of capillary waves with a wave vector κ

$$\tilde{t}_1 \cong \left(\frac{\rho}{\gamma\kappa^3} \right)^{1/2}.$$

In all what follows, we shall be concerned only with the high viscosity regime ($\tilde{t}_1 < t_1$).

3. The dynamic creeping film.

3.1 A REMINDER ON HORIZONTAL SPREADING. — When a wetting fluid spreads over a horizontal solid, with a fixed velocity U (Fig. 2) the macroscopic contact line ζ_0 is preceded by a precursor film, first discovered by Hardy [21]; the structure of this precursor has been investigated experimentally [22] and theoretically [23-25]. Many situations are possible : again it is helpful to focus on the relatively simple case of a Van der Waals fluid, where results can be written down in simple terms [24-25]

a) a film, or « tongue », shows up when the dynamic contact angle θ has reached low values

$$\theta < \phi \equiv \left(\frac{S}{\gamma} \right)^{1/2}; \quad (11)$$

b) when $\theta \ll \phi$ the thickness of the film at the cross-over point is

$$\zeta_0 \sim a/\theta; \quad (12)$$

c) the film is truncated at a thickness $e = e_s$ where e_s has been defined in equation (2). For Van der Waals fluids :

$$e_s = a \left(\frac{3\gamma}{2S} \right)^{1/2} \quad (13)$$

$$\cong a\phi^{-1}; \quad (14)$$

d) the extension of the tongue (from the point $e = \zeta_0$ to the tip $e = e_s$) is

$$l \cong a^2 \theta^{-3}/e_s; \quad (15)$$

⁽¹⁾ We are indebted to a referee for this observation.

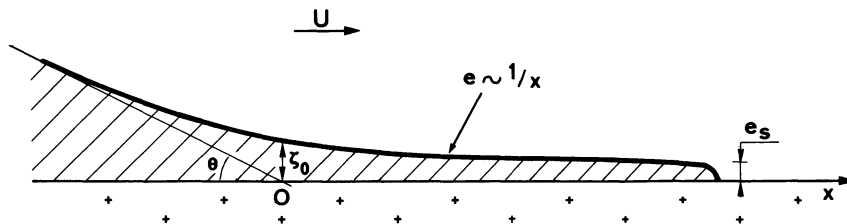


Fig. 2. — The « adiabatic » precursor for a Van der Waals fluid spreading on a solid, with a fixed velocity U (after Refs. [16], [24]). The film is truncated at a thickness e_s (Eq. (14)).

e) the velocity U of the whole pattern is related to the angle θ by the Hoffmann-Tanner law

$$U \cong V^* \theta^3. \quad (16)$$

A qualitative understanding of equation (15) can be obtained in terms of a *diffusion coefficient* $D(e)$ for a flat film of thickness e [25]. Balancing disjoining pressures against viscous forces, one finds that a small perturbation of the thickness decays according to a diffusion equation, with a coefficient

$$D(e) = -\frac{e^3}{3\eta} \frac{d\Pi}{de}. \quad (17)$$

For Van der Waals fluids

$$D(e) = \frac{A}{6\pi\eta e} = \frac{V^* a^2}{e}. \quad (18)$$

An essential feature is that $D(e)$ becomes large at small thicknesses. The tongue size l of equation (15) can be understood in terms of a competition between diffusion at the tip ($D(e_s)$) and drift (velocity U)

$$l = D(e_s)/U. \quad (19)$$

3.2 FINE STRUCTURE OF THE «TONGUE». — The above discussion assumed that the whole fluid pattern was drifting with a constant velocity U with respect to the solid. This is strictly correct for the dipping problem of reference [12]. But for transient problems, such as the one considered here, the situation may be more complex. It is described qualitatively on figure 3. We have a nominal contact line \mathcal{L}_0 moving at a velocity $U(t)$, and we use a reference frame where the solid plate moves at the velocity $-U$.

a) Starting from \mathcal{L}_0 (at $x = 0$) we find a film which has reached a *steady state* in a certain interval ($0 < x < x_1$). Here the current J vanishes, and this gives

$$J = -eU - D(e) \frac{\partial e}{\partial x} = 0 \quad (20)$$

from which we can see that the scaling form of the profile is given by the implicit equation

$$x = D(e)/U \quad (21)$$

a generalization of (19). In particular, with VdW forces, the result is $e \sim x^{-1}$ as discussed in reference [16]. The film present in the interval ($0 < x < x_1$) will be called the « *adiabatic* » film.

b) At larger distances ($x_1 < x < l$) we have a *diffusive* regime, where the drift term (U) becomes negligible, and where the transport equation reduces mainly to

$$\frac{\partial e}{\partial t} = \frac{\partial}{\partial x} \left(D(e) \frac{\partial e}{\partial x} \right). \quad (22)$$

Here, the scaling law for the profile is

$$x^2 = 2 D(e) t. \quad (23)$$

In particular, the tip of the tongue (thickness e_s) is associated with the fastest diffusion coefficient $D(e_s)$.

c) The crossover point ($x = x_1$) between the adiabatic film and the diffusive film, corresponds to

$$x_1 \cong Ut \quad (24)$$

$$e(x_1) = e_1 = \frac{a^2 V^*}{U^2 t}. \quad (25)$$

In equation (25) we have gone to Van der Waals fluids and used equation (18) for $D(e)$. Of course, (25) will be meaningful when the thickness e_1 is in the right interval

$$e_s < e_1 < \zeta_0.$$

On the other hand, if $e_1 < e_s$, the whole tongue is adiabatic and equation (19) holds.

3.3 APPARITION OF A DYNAMIC PRECURSOR IN VERTICAL CREEP. — We now return to the climbing problem of figure 1, and transpose the results :

a) A film shows up at a time t_2 such that $\theta(t_2) = \phi$ (Eq. (12)). Inserting the form (8) for $\theta(\tau)$ we find

$$\tau_2 = \frac{\gamma}{S} \equiv \phi^{-2}. \quad (26)$$

With our choice of values ($S/\gamma = 10^{-2}$) we arrive at times $t_2 \sim 10^{-2}$ s.

b) At times $t > t_2$ (but below a certain limit t_3 to be defined below) we expect a purely adiabatic « tongue » with a crossover thickness derived from equation (12).

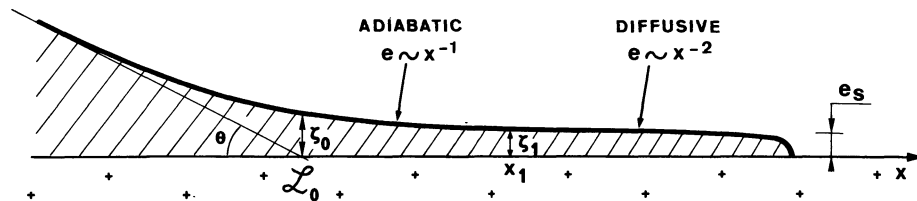


Fig. 3. — A « mixed » precursor — in the region from $x = 0$ to $x = x_1$ the velocity U of the nominal contact line \mathcal{L}_0 imposes the average fluid velocity. Beyond x_1 , the flow is controlled by non linear diffusion.

$$\zeta_0 = a\theta^{-1} = a\tau^{1/2}. \quad (27)$$

The length l of the tongue is given by equations (19) and (7, 8)

$$\kappa l(t) = \varepsilon \phi \tau^{3/2}. \quad (28)$$

c) At a certain time t_3 the tongue becomes diffusive. We may construct t_3 through the crossover thickness e_1 defined in equation (25). Inserting equations (7, 8) into (25) we arrive at

$$\kappa e_1 = \varepsilon^2 \tau^2. \quad (29)$$

The tip of the tongue becomes diffusive when e_1 increases up to the tip thickness e_s (Eq. (14)). Equating (29) and (14) we find the corresponding crossover time

$$\tau_3 = (\varepsilon \phi)^{-1/2}. \quad (30)$$

Note that $\tau_3/\tau_2 \sim (\phi^3 \varepsilon)^{1/2}$ is larger than unity because of our starting assumptions. For our numerical example $t_3 \sim 1$ s.

At times $t \gg t_3$ the tongue is mainly diffusive, and has a length derived from equation (23), namely $l^2 \cong D(e_s) t$, or :

$$\kappa l = (\phi \varepsilon \tau)^{1/2}. \quad (31)$$

Our reader may check directly that the forms (28) and (31) do crossover at $\tau = \tau_3$.

d) Ultimately, at a certain time t_4 , two things will happen to the « tongue » :

(i) the adiabatic portion fades out : the crossover thickness e_1 becomes comparable to ζ_0 . Comparing equations (25) and (27) this leads to

$$\tau_4 = \varepsilon^{-2/3}; \quad (32)$$

(ii) a piece of static film (described by Eq. (5)) appears. This corresponds to $\zeta_0 = e_0(h_c)$ and also leads to equation (32). The build up of the static film will be analysed in the next section.

A final remark concerning τ_4 : we can check that

$$\tau_4/\tau_3 = (\phi^3/\varepsilon)^{1/6} > 1$$

with our assumptions on ϕ . Numerically, for our particular choice of values, the time t_4 is of order 10 seconds.

4. Development of the static film.

4.1 THE DIFFUSION APPROXIMATION. — After time t_4 , a static film begins to build up above the macroscopic meniscus. The upper limit of this static portion is a certain altitude $h_i(t)$ (Fig. 1). Diffusion signals spread out from the meniscus ($h = h_c$) and sweep all the region up to h_i . These signals are carried with the fastest diffusion coefficient available, namely

$$D_1 = D(e_0(h_i)) \cong \frac{V^* a^2}{e_0(h_i)} \quad (33)$$

since $e_0(h_i)$ is the minimum thickness in this region. This leads to a self-consistent formula for h_i :

$$\Delta^2 \equiv (h_i - h_c)^2 \cong D_1(t - t_4) \sim D_1 t \quad (t_4 \ll t). \quad (34)$$

a) At times $t < t_5$ we have $\Delta < h_i$ and then the argument in D_1 may be treated as constant

$$D_1 \rightarrow D_c \equiv \frac{V^* a^2}{e_c} = V^* a \varepsilon^{1/3}. \quad (35)$$

We then have a case of simple diffusion ($\Delta \sim t^{1/2}$). More precisely :

$$\kappa \Delta = \tau^{1/2} \varepsilon^{2/3} = (\tau/\tau_5)^{1/2} \quad (\tau < \tau_5) \quad (36)$$

$$\tau_5 = \varepsilon^{-4/3}. \quad (37)$$

The time t_5 is typically of order 10 days !

b) If we insist on probing longer times ($t \gg t_5$) then h_c becomes negligible with respect to h_i in equation (34). The self-consistent solution is then :

$$\kappa h_i(\tau) \cong \kappa \Delta(\tau) \cong (\tau/\tau_5)^{3/5}. \quad (38)$$

This scaling law is the exact analog of what we had in dipping a plate at ultralow velocities [9]. Equation (38) can in fact be derived by seeking suitable self-similar solutions, as shown in the appendix.

c) To be sure that the diffusion approximation (Eq. (34)) was the right starting point, we must check that the film is diffusive rather than adiabatic. The ratio $\Delta(t)/(Ut)$ (where U is the drift velocity defined in equation (7)) must be larger than unity. Indeed, with good use of equations (7, 8, 36) we find that

$$\frac{\Delta(t)}{Ut} = \frac{\tau}{\tau_4} > 1 \quad (\tau_4 < \tau < \tau_5) \quad (39)$$

and thus, whenever $\tau > \tau_4$, the regime is purely diffusive.

d) Finally we should mention the tongue — here, as soon as we passed the time t_3 , we expect the diffusion law (31) to hold — the tip has always a thickness e_s and moves with the fastest available diffusion constant $D(e_s)$.

An attentive reader will note that the static portion, described by $h_i(t)$ (Eq. (38)) ultimately grows faster than the tongue, and must catch it up at some point ! this is no surprise — comparing (38) and (31) we find that the two things merge when we have reached the ultimate permissible size h_m , defined by the dewetting effect of the gravitational potential (Eq. (2)). But the time required to reach h_m is astronomically large, and not physically significant.

5. Conclusions.

Our scaling analysis does predict a remarkable cascade of events for the wetting of a vertical wall by a non volatile Van der Waals fluid. The first stage ($t \sim t_1$)

depends on the exact initial conditions and is not interesting. The birth of a precursor at time t_2 is not a great surprise — it is very similar to what we expect for the horizontal spreading of a droplet. The transition between « adiabatic » and « diffusive » precursors, occurring at a later time t_3 , is also expected for horizontal spreading, but had not been discussed yet (to our knowledge). The onset of a static film (at time t_4) is a specific feature of the vertical geometry, and deserves experimental studies. The final growth (at times $t > t_5$) is ruled by a nice self-similar type of solution but experiments over such long times appear difficult, because of various problems (contamination, temperature stability, ...). It is interesting to see, however, that so many things should happen in the interval from one second to one day, which is most relevant for soil physics.

Various extensions of the present work are clearly needed :

a) toward mathematical rigor — many crossovers are involved; fortunately they are rather well separated in the time sequence and in space; thus they could be studied separately by suitable matching of asymptotic expansions;

b) towards more practical fluids, such as water and also towards two-fluid systems (water replacing oil);

c) possibly allowing for exchanges through the vapour;

d) investigating the stability of the film fronts with respect to small perturbations.

But, at this moment, a first experimental program on simple Van der Waals fluids could be started with some guidance from the scaling laws, using various optical methods to monitor the films.

APPENDIX

Self-similar film profile.

In the late stages of the creep ($t > t_5$), the profile of the ascending film can be described in more details by looking for self-similar solutions to the dynamical equations.

In the lubrication approximation, at a height h over the reservoir level, the thickness of the film is related to the local upward velocity U by

$$\eta U = e^2 \left[-\rho g/3 - \frac{A}{6\pi e^4} \frac{\partial e}{\partial h} \right]. \quad (\text{A.1})$$

The first term on the right hand side represents the gravity force, the second is the disjoining pressure gradient for a Van der Waals fluid. For heights much larger than the capillary length h_c , the film is almost flat parallel to the solid surface and it is legitimate to neglect the Laplace pressure.

The other relation between velocity and thickness is provided by the conservation equation

$$\frac{\partial e}{\partial t} + \frac{\partial e}{\partial x} U = 0. \quad (\text{A.2})$$

Combining these two equations we get the profile equation

$$-\frac{\rho g}{\eta} e^2 \frac{\partial e}{\partial h} - \frac{\partial}{\partial h} \left[\frac{A}{6\pi\eta e} \frac{\partial e}{\partial h} \right] + \frac{\partial \zeta}{\partial t} = 0. \quad (\text{A.3})$$

In the absence of gravity ($g = 0$) this is a non linear diffusion equation with a thickness-dependent diffusion coefficient

$$D(e) = \frac{A}{6\pi\eta e}.$$

In the microscopic regime there is just one characteristic height $h_1(t)$ for the film given by equation (38). It is then reasonable to look for a self similar solution to equation (A.3) in the form

$$e = e(h_1) f\left(\frac{h}{h_1}\right).$$

The function f satisfies then the dimensionless differential equation

$$\frac{df}{du} \left[f^2 + \frac{3}{5}u \right] + \frac{d}{du} \left[\frac{1}{f} \frac{df}{du} \right] + f/5 = 0. \quad (\text{A.4})$$

In order to discuss the solutions of this equation, we distinguish 2 regions in the microscopic film :

i) at small heights, $u = h/h_1 \ll 1$ (h being however larger than h_1) the static film is already established; the viscous terms are then negligible in equation (A.4), which reads :

$$f^2 \frac{df}{du} + \frac{d}{du} \left[\frac{1}{f} \frac{df}{du} \right] = 0$$

f is thus given by the usual static solution (Eq. (5))

$$f(u) = u^{-1/3};$$

ii) at larger heights the film is not established yet, it is thus much thinner than its equilibrium value. The gravity term is negligible compared to the Van der Waals disjoining pressure

$$\frac{3u}{5} \frac{df}{du} + \frac{f}{5} + \frac{d}{du} \left(\frac{1}{f} \frac{df}{du} \right) = 0.$$

The film profile is thus

$$f(u) = \frac{2}{u^2} \quad (\text{A.5})$$

or in the original units

$$e(h, t) = 2 V^* t \frac{a^2}{h^2}. \quad (\text{A.6})$$

In practice, the large height profile is valid over a much larger time range than studied here. As soon as $t > t_3$ when the dynamic precursor film appears, there exists a region where the gravity force can be neglected and where the profile can be approximated by (A.6). This film is cut at a thickness $e(l, t) = e_s$

leading back to the tongue length given by equation (24)

$$l(t) = \left[2 V^* t \frac{a^2}{e_s} \right]^{1/2}.$$

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