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ON THE BEHAVIOUR OF A THIN LIQUID LAYER FLOWING DUE TO GRAVITY AND A SURFACE TENSION GRADIENT

C.I. GHEORGHIU

Dedicated to Professor Ioan A. Rus on his 60^{th} *anniversary*

Abstract. A thin liquid layer flowing due to gravity and a surface tension gradient is taken into account. On the liquid/gas interface one of the boundary conditions reduces to the fact that the normal stress equals the atmospheric pressure. This is the main difference between our study and those where the same boundary condition expresses the fact that the normal stress is proportional to the curvature. In these, by using the standard lubrication theory, a fourth-order nonlinear parabolic equation for the fluid film height is obtained. In ours, by using the same theory, a nonlinear conservation law with a nonconvex flux function is deduced for the same variable. For this equation a similarity solution is carried out. It shows that the behaviour of the liquid layer depends essentially upon the gradient of surface tension and is quite insensitive to the viscosity of the liquid. "Viscous" and weak formulations for the conservation law are also carried out. An entropy condition to pick out physically relevant weak solutions is used.

1. Introduction

The thin film theory (lubrication theory) and similarity methods are used to determine the behaviour of the free surface (the liquid / gas interface) of a thin liquid layer flowing due to gravity and a gradient of surface tension. This gradient acts on the liquid / gas interface (the upper surface of the liquid layer). The surface tension σ at each point of the interface is related to the local surfactant concentration Γ through an empirically determined equation of state $\sigma(\Gamma(x))$. The gradient in Γ, and thus in σ, along the interface induces a shear stress at the surface of the underlying liquid, and thus a Marangoni flow in the substrate. If the liquid substrate is thin, and if diffusion of the surfactant on the surface of the layer is sufficiently slow, and consequently negligible, that shear stress induces large deformations in the layer of liquid. From the mathematical point of view this gradient of surface tension behaves like an advancing rigid plate. Thus, if the initial

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gradients in surface tension are sufficiently large, the deformations of the liquid layer may be severe enough leading the film to rupture.

In order to refine the similarity solutions, a "viscous" and also a weak equation for the evolution of the interface $z = h(x,t)$ are deduced.To peak out the physically relevant weak solution an entropy condition is displayed. Numerical solutions starting from both "viscous" and weak formulations will be the aim of a following work.

The dynamics of thin liquid layers is important in many industrial process, from painting a car-body to coating a microchip ([5], [6]) and also in medicine in the development of the respiratory distress syndrome of many prematurely born infants ([3], [6] and [7]).

The last two quoted works represent a very keen analysis on the existence of shock profiles. They also give a continuous dependence result for the initial value problem encountered in flows described above.

Our analysis is eventually orientated towards numerical results.

2. The model

The model to be investigated here has been described in details in our previous work [2] and accordingly only a brief summary is given here. We will consider a thin liquid layer of a viscous incompressible Newtonian fluid flowing on a rigid inclined plane (α is the slope). A monolayer of insoluble surfactant creates a gradient of surface tension which acts at the upper surface of the layer. Thus, this gradient of surface tension can act along or against gravity.

The variables of the flow are scaled as follows. Let U be a typical velocity corresponding to undisturbed height d of the layer. We consider $U = \rho g d^2 \sin\alpha/\mu$ as the average velocity of the undisturbed flow, where ρ is the density assumed constant, g gravitational acceleration and μ is fluid's dynamic viscosity. According to what we reported in [2], the aspect ratio $\varepsilon = d/L \ll 1$, where L is the initial length of the layer, thus the thin film theory ([1], p.239), may be used.

From the equation of mass conservation we are led to scale the vertical velocity by $\varepsilon\, U$. We choose to scale time by $h/\varepsilon\, U$ and the pressure by $\rho\, U^2$. We also suppose that the Reynolds number $Re = \rho U d/\mu$ is sufficiently small so that the leading order inertial terms in momentum equation, of $O(\varepsilon^2 Re)$, are negligible.

The surface tension σ at each point of monolayer is related to the local surfactant concentration Γ through an empirically determined equation of state $\sigma = \sigma(\Gamma(x))$. The gradient in Γ, and thus in σ, along the monolayer induces a shear stress at the surface of underlying liquid, and thus a Marangoni flow in the substrate. If the liquid substrate is thin (as we assume), and if diffusion of the surfactant on the upper surface of the layer is sufficiently slow, the flow induces large deformations in the layer (Jensen & Grotberg [3]).

Neglecting surface diffusivity of the surfactant, our aim is to analyse these deformations.

A very elaborate discussion on the dependence of σ on Γ can be found in [3]. In the expression of the gradient of surface tension $d\sigma/dx = \frac{d\sigma}{d\Gamma}\frac{d\Gamma}{dx}$, $d\sigma/d\Gamma$ is in general nonlinear, although a linear law is predominantly used in literature. Our analysis remains chiefly qualitative and mathematically orientated so we do not pay more attention to these aspects.

If σ is scaled by σ_0, the higher surface tension on the liquid / gas interface, and if we take the coordinates (x, z), with z vertical to plane and x downwards the plane, scaled by d, the corresponding velocity field is $(u(x,z,t), w(x,z,t))$. The upper surface of the layer is at $z = h(x,t)$.

We notice that in practice it is highly unlikely that gravitational and intermolecular forces (van der Waals forces) would ever be of the same order. As in the work of Jensen & Grothberg [3], the influence of intermolecular forces is deeply analysed, our intention is to concentrate on the dependence of the behaviour (deformations) of the liquid layer upon the competition between gravity and the surface tension gradient.

Thus, the equations of momentum and mass conservation for the layer of liquid are

$$0 = -p_x + \frac{1}{Re}u_{zz} + \frac{\sin\alpha}{F^2} \tag{1}$$

$$0 = -p_z - \frac{\cos\alpha}{F^2} \tag{2}$$

$$u_x + w_z = 0 \tag{3}$$

where $F = U/(gh)^{1/2}$ is the Froude number and subscripts denote differentiation with respect to that variable.

On integrating the second of these,

$$p = -\frac{\cos\alpha}{F^2} z + f(x,t).$$

On the liquid / gas interface, $z = h(x,t)$, the condition that the normal stress be equal to atmospheric pressure p_0 reduces essentially to $p = p_0$, so

$$p(x,z,t) = \frac{\cos\alpha}{F^2}[h(x,t) - z] + p_0 \tag{4}$$

On this boundary condition we will comment at the end of the paper.

The tangential stress condition at $z = h$ reads

$$u_z = Ca \frac{d\sigma}{d\Gamma}\frac{d\Gamma}{dx} \tag{5}$$

where $Ca = \sigma_0/U\mu$ is the capillary number.

With (2.4) the equation of motion (2.1) becomes

$$\frac{1}{Re} u_{zz} = -\frac{\sin\alpha}{F^2} + \frac{\cos\alpha}{F^2} h_x.$$

Now, h_x is small, by virtue of the thin film approximation. Thus, unless α is very small, the last term may be neglected and with the boundary condition (2.5) and the no - slip condition at $z = 0$, we find

$$u = -\frac{z^2}{2} + \left(h + Ca\frac{d\sigma}{d\Gamma}\frac{d\Gamma}{dx}\right) z. \tag{6}$$

Taking into account that the quantities $d\sigma/d\Gamma$ and $d\Gamma/dx$ are given, the incompressibility condition (2.3) gives

$$w_z = -u_x = -z\, h_x.$$

On integration and application again of the no - slip boundary condition we have

$$w = -\frac{z^2}{2} h_x. \tag{7}$$

The final consideration is the purely kinematic condition at the free surface. In dimensionless form it reads:

$$w = \varepsilon h_t + u h_x,$$

or with u and w form (2.6), (2.7)

$$\varepsilon\, h_t + \left(h^2 + Ca\frac{d\sigma}{d\Gamma}\frac{d\Gamma}{dx}h\right) h_x = 0. \tag{8}$$

The evolution equation for $h(x,t)$ is therefore (2.8).

3. Similarity and weak solutions

The solution of this last equation is by similarity

$$h = f\left[x - \frac{1}{\varepsilon}\left(h^2 + Ca\frac{d\sigma}{d\Gamma}\frac{d\Gamma}{dx}h\right)t\right] \tag{9}$$

where f is an arbitrary function of a single variable, so any particular value of h propagates up or down with speed

$$h^2 + Ca\frac{d\sigma}{d\Gamma}\frac{d\Gamma}{dx}h \tag{10}$$

depending on the sign of this quantity. Thus for some negative values of the gradient of surface tension, waves travelling upward are possible. This fact is in accordance with our observations reported in [2] and the works quoted there. Plainly, h remains constant if $x - \frac{1}{\varepsilon}\left(h^2 + Ca\frac{d\sigma}{d\Gamma}\frac{d\Gamma}{dx}h\right)t$ does. The function f could be determined by adding to (2.8) appropriate initial conditions.

If no gradient of surface tension acts ($d\sigma/dx = 0$), from (3.1) the results of Acheson [1], p.247, are confirmed.

Due to the form of (3.2) an explicit similarity solution of (2.8) writes

$$h = \frac{-Ca\frac{d\sigma}{dx} \pm \sqrt{(Ca\sigma_x)^2 + 4\frac{\varepsilon x}{t}}}{2}. \tag{11}$$

Thus, as time goes on, the main part of the perturbation, denoted by h, approaches this simple similarity solution more or less depending upon the initial conditions.

However, the evolution equation for h, (2.8), can be written as a nonlinear conservation law ([4]):

$$h_t + F(h)_x = 0, \tag{12}$$

where the flux function

$$F(h) = \frac{1}{\varepsilon}h^3 + \frac{1}{2}Ca\sigma_x h^2$$

is a nonconvex one.

The corresponding "viscous" equation for (3.4) reads as follows:

$$h_t + F(h)_x = \gamma h_{xx}, \quad 0 < \gamma \ll 1. \tag{13}$$

Here γ is the "viscous" parameter and a solution of this, for vanishing γ, is called an entropy solution or a vanishing viscosity solution.

A wealk formulation for (3.4) is obtained in a straightforward manner. Multiplying the equation by a smooth "test function" $\Phi \in C_0^1(\mathbf{R} \times \mathbf{R})$ - the space of functions that are continuously differentiable with "compact support", we obtain the problem:

$$\begin{cases} \text{find } u \in L^3(\mathbf{R} \times \mathbf{R}) \text{ such that} \\ \int\limits_0^\infty \int\limits_{-\infty}^\infty [u\Phi_t + F(u)\Phi_x]\, dtdx = -\int\limits_{-\infty}^\infty \Phi(x,0)\, u(x,0)\, dx, \forall \Phi \in C_0^1(\mathbf{R} \times \mathbf{R}). \end{cases} \tag{14}$$

Thus a solution of (3.6), named a weak solution, if it exists, involves no derivative on u and hence requires less smoothness than the corresponding solutions of the "viscous" equation (3.5) or even "inviscid" equation (2.8). Unfortunately, weak solutions are often not unique, and so an additional question is to identify which weak solution is the physically correct vanishing viscosity solution. In order to avoid working with the "viscous" equation directly, we will formulate another condition on weak solutions which is easier to check, and which will also pick out the physically relevant solutions. This is the so called entropy condition (due to Oleinik, [4], p.36) which reads as follows: $h(x,t)$ is the entropy solution if all discontinuities propagating with speed s given by $F(h_l) - F(h_r) = s(h_l - h_r)$ have the property that

$$\frac{F(h) - F(h_l)}{h - h_l} \geq s \geq \frac{F(h) - F(h_r)}{h - h_r} \tag{15}$$

for all h between h_l and h_r.

Finally we observe that the case of F nonconvex is more complicated mathematically than that of F convex and more important the entropy solution might involve both a shock or a rarefaction wave.

4. Concluding remarks

The similarity solution (3.3) can be interpreted as follows:

for $t \to \infty$,

$$h = \begin{cases} |Ca\sigma_x|, & \sigma_x < 0 \\ 0, & \sigma_x \geq 0. \end{cases}$$

This means that a negative gradient of surface tension could sustain a liquid layer of height $|Ca\sigma_x|$ and a positive gradient does not. It is phisically plausible and is in fact a linear theory of thin liquid layer (thin liquid film) rupture. Moreover, this result is quite insensitive to the viscosity of the liquid. It depends essentially upon the sign of the gradient of surface tension, confirming the fact that this gradient drives the system. In [9] one could find a nonlinear theory of film rupture for a horizontal liquid film. There the surface tension is assumed to be constant, London / van der Waals forces are included, but double - layer forces are neglected.

The "viscous" equation for evolution equation, (3.5), and the weak formulation of that, (3.6), with entropy condition, (3.7), create a fine background on which numerical methods could work. Such numerical results could refine the rough information given by similarity solution.

They will be the aim of a following paper.

On the importance of the boundary condition for pressure on the liquid/gas interface we have the following comment. If one take the Laplace-Young equation (the normal stress due to surface tension is proportional to curvature) as a boundary condition, instead of $p = p_0$, $z = h$, which is physically motivated, he obtains an equation for $h(x,t)$ which is similar to the lubrication one from [5]. It reads:

$$\varepsilon h_t + \frac{1}{3}\left[h^3\left(Reh_{xxx}+1\right)+\frac{1}{2}Ca\sigma_x h^2\right]_x = 0.$$

A proper comparison between these two type of boundary conditions and their implications on the theory of flows where the surface tension is a driving mechanism remains an open problem.

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INSTITUTE OF MATHEMATICS, CLUJ-NAPOCA, ROMANIA