

LETTERS

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Stokes-flow instability due to interfacial surfactant

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The linear stability of a two-fluid shear flow with an insoluble surfactant on the flat interface is investigated in the Stokes approximation. Gravity is neglected in order to isolate the Marangoni effect of the surfactant. In contrast to all earlier studies of related fluid systems, we encounter (i) the destabilization (here, of a shear flow) caused solely by the introduction of an interfacial surfactant and (ii) the destabilization (here, of a system with a surfactant) caused solely by the imposition of a Stokes flow. Asymptotic long-wave expressions for the growth rates are obtained. © 2002 American Institute of Physics. [DOI: 10.1063/1.1483838]

Among instabilities of viscous two-fluid systems,¹ the Yih^2 instability—driven by the interfacial jump in viscosity—occurs only if a basic (shear) flow is present. However, the Yih instability depends on inertial terms of the Navier–Stokes equations. Instabilities which exist for “inertialess,” Stokes flows, do not disappear if the basic flow is turned off. Examples of such Stokes flow instabilities include the gravitational Rayleigh–Taylor (RT) instability due to the different densities in a horizontally extended two-fluid system (e.g., such as in Ref. 3) and the capillary Rayleigh instability of the core-annular systems (as in Ref. 4).

In the present Letter, we demonstrate what we believe is the first example of a Stokes flow instability which does disappear if the basic flow is stopped. For a range of two-layer plane Couette–Poiseuille flows, it turns out that if there is an insoluble surfactant present at the interface and the surface tension depends on the surfactant concentration, the flow is unstable to sufficiently long waves. However, the system is stable to small disturbances of any wavelength if the basic flow is absent. Perhaps even more surprisingly, we encounter cases of stable surfactant-free flows which become unstable if an interfacial surfactant is introduced.

There has been considerable interest in the instability of multifluid film flows and the influence of surfactants since they occur in many industrial and biomedical applications—such as lubricated pipelining,¹ coating in photography,⁵ and the obstruction to airflow in the small airways of the lungs.^{6,7} The effect of insoluble surfactant on stagnant film systems was always purely stabilizing. (See, e.g., Refs. 8–10. For a broader survey of multifluid instabilities, see, e.g., the recent papers Refs. 11 and 12.)

In this paper, we investigate the stability of two-layer plane Couette–Poiseuille flow between two parallel plates with one plate moving steadily, in a possible presence of a uniform pressure gradient, and with an insoluble surfactant monolayer on the interface between the two fluid layers. A

case of inertialess, Stokes flow, is considered with no external forces (such as gravity, molecular van der Waals forces, etc.), so that if the surface tension is assumed constant, the system is stable for all wave numbers. (For this, the plane geometry is essential: in the core-annular geometry, even a constant surface tension can be destabilizing due to azimuthal curvature of the interface.) Thus, in our model, the instability is due solely to the interaction between the basic flow and the surfactant monolayer.

The hints to an instability due to surfactant can be already seen in the recent work by Wei and Rumschitzki.¹³ However, they considered a core-annular flow, in which case the instability is mainly due to the curvature effects mentioned above. Only a correction to that main instability is due to the surfactant; in contrast, in our case the surfactant is the sole agent driving the instability.

The exact formulation of the problem is as follows. Consider two immiscible fluid layers between two infinite parallel plates, as in Fig. 1 of Ref. 2. Let the basic flow be driven by the combined action of an in-plane steady motion of one of the plates and a constant pressure gradient parallel to the plate velocity. It is well known that the basic “Couette–Poiseuille” velocity profiles are steady and vary (quadratically) in the spanwise direction only, and the basic interface between the fluids is flat. For simplicity, let the densities of the two fluids be equal. Then gravity does not affect stability of the basic flow, and is disregarded below. It is convenient to use the reference frame of the unperturbed interface. Let y^* be the spanwise, “vertical,” coordinate (the symbol $*$ indicates a dimensional quantity). Let the interface be at $y^* = 0$ and the y^* -axis directed from the thinner layer to the thicker one; we will call this the “upward” direction (clearly, since there is no gravity, the notions of “up” and “down” are a matter of convention). Thus, $d_1 < d_2$ holds, where d_1 and d_2 are the thicknesses of the lower and upper fluids, respectively. The direction of the “horizontal” x^* -axis is chosen so

that velocity of the lower plate, situated at $y^* = -d_1$, is $-U_1$ if U_1 is the relative speed of the interface and the lower plate. The velocity U_2 of the upper plate (situated at $y^* = d_2$) is positive for the purely Couette flow; however, it does not have to be positive in the presence of a pressure gradient. For the Couette flow, in which the velocity profiles are linear, it is easy to see that, in terms of U , the velocity of the upper plate relative to the lower plate (i.e., $U = U_1 + U_2$), we have $U_1 = \mu_2 d_1 (\mu_2 d_1 + \mu_1 d_2)^{-1} U$ and $U_2 = \mu_1 d_2 (\mu_2 d_1 + \mu_1 d_2)^{-1} U$, where μ_1 and μ_2 are the viscosities of the lower and upper fluids, respectively. For the more general (quadratic) Couette–Poiseuille flow, it is also not difficult to express U_1 and U_2 in terms of the two “physical” quantities, U and the basic pressure gradient; however, we will not need these expressions.

As in Yih,² the well-known Squire’s theorem allows us to confine our consideration to two-dimensional perturbed flows (in the x^*y^* -plane). The equation of the perturbed interface is $y^* = \eta^*(x^*, t^*)$, and the Navier–Stokes and incompressibility equations governing the fluid motion in the two layers are (with $j=1$ for the lower layer and $j=2$ for the upper one)

$$\rho \left(\frac{\partial \mathbf{v}_j^*}{\partial t^*} + \mathbf{v}_j^* \cdot \nabla^* \mathbf{v}_j^* \right) = -\nabla^* p_j^* + \mu_j \nabla^{*2} \mathbf{v}_j^*, \quad \nabla^* \cdot \mathbf{v}_j^* = 0, \quad (1)$$

where $\nabla^* = (\partial/\partial x^*, \partial/\partial y^*)$, ρ is the density (of both fluids), $\mathbf{v}_j^* = (u_j^*, v_j^*)$ is the fluid velocity with horizontal component u_j^* and vertical component v_j^* , and p_j^* is the pressure.

We use the “no-slip, no-penetration” boundary conditions (requiring zero relative velocities) at the plates: $u_1^* = -U_1$, $v_1^* = 0$ at $y^* = -d_1$; and $u_2^* = U_2$, $v_2^* = 0$ at $y^* = d_2$. The interfacial boundary conditions are as follows. The velocity must be continuous at the interface: $[\mathbf{v}^*]_1 = 0$, where $[A]_1^2 = A_2 - A_1$ denotes the jump in A across the interface, i.e., at $y^* = \eta^*(x^*, t^*)$. The interfacial balances of the tangential and normal stresses taking into account the gradient of surface tension and the capillary jump in the normal stress are

$$\frac{1}{1 + \eta_{x^*}^{*2}} [(1 - \eta_{x^*}^{*2}) \mu (u_{y^*}^* + v_{x^*}^*) + 2 \eta_{x^*}^{*2} \mu (v_{y^*}^* - u_{x^*}^*)]_1^2 = - \frac{\sigma_{x^*}^*}{(1 + \eta_{x^*}^{*2})^{1/2}}, \quad (2)$$

$$[(1 + \eta_{x^*}^{*2}) p^* - 2 \mu (\eta_{x^*}^{*2} u_{x^*}^* - \eta_{x^*}^* (u_{y^*}^* + v_{x^*}^*) + v_{y^*}^*)]_1^2 = \frac{\eta_{x^* x^*}^*}{(1 + \eta_{x^*}^{*2})^{3/2}} \sigma^*, \quad (3)$$

where σ^* is the surface tension. The kinematic interfacial condition is $\eta_{t^*}^* = v^* - u^* \eta_{x^*}^*$. The surface concentration of the insoluble surfactant on the interface, Γ^* , obeys an equation (see, e.g., Ref. 14) which for the one-dimensional case becomes

$$\frac{\partial \Gamma^*}{\partial t^*} + \frac{\partial}{\partial s^*} (\Gamma^* u_s^*) + \Gamma^* \kappa^* u_n^* = D_s \frac{\partial^2 \Gamma^*}{\partial s^{*2}}, \quad (4)$$

where s^* is the arclength along the interface (so that $\partial/\partial s^* = (1/\sqrt{1 + \eta_{x^*}^{*2}})(\partial/\partial x^*)$), u_s^* and u_n^* are the tangential and normal components of the surface velocity, $\kappa^* = \eta_{x^* x^*}^*/(1 + \eta_{x^*}^{*2})^{3/2}$ is the interfacial curvature, and D_s is the surface molecular diffusivity of surfactant; D_s is usually negligible and is discarded below. Since we only deal with infinitesimal deviations of the concentration Γ^* from its basic value Γ_0 , we can linearize the surface tension dependence on the surfactant concentration: $\sigma^* = \sigma_0 - E(\Gamma^* - \Gamma_0)$, where σ_0 is the basic surface tension and E is a constant.

We introduce dimensionless quantities as follows: $(x, y) = (x^*, y^*)/d_1$, $t = t^*/(d_1 \mu_1 / \sigma_0)$, $(u, v) = (u^*, v^*)/(\sigma_0 / \mu_1)$, $p = p^*/(\sigma_0 / d_1)$, $\Gamma = \Gamma^*/\Gamma_0$, and $\sigma = \sigma^*/\sigma_0$. The dimensionless velocity field of the basic Couette–Poiseuille flow, with a flat interface, $\eta = 0$, and uniform concentration of surfactant, $\bar{\Gamma} = 1$ (where the overbar indicates a basic-state quantity), is $\bar{v}_1 = 0$, $\bar{u}_1(y) = sy + qy^2$ (for $-1 \leq y \leq 0$) and $\bar{u}_2(y) = \bar{u}_1(y)/m$, $\bar{v}_2 = 0$ (for $0 \leq y \leq n$, where $1 \leq n = d_2/d_1$ and $m = \mu_2/\mu_1$). The constants s and q will be used in place of the pressure gradient and the relative velocity of the plates to characterize the basic flow. As will be seen below, the stability depends only on the coefficient s , the shear of the basic velocity at the interface: $s = D\bar{u}_1(0)$, where $D = d/dy$.

We consider the perturbed state with small deviations from the basic flow: $\eta = \tilde{\eta}$, $u_j = \bar{u}_j + \tilde{u}_j$, $v_j = \bar{v}_j + \tilde{v}_j$, $p_j = \bar{p}_j + \tilde{p}_j$ and $\Gamma = \bar{\Gamma} + \tilde{\Gamma}$. It is convenient to introduce disturbance streamfunctions $\tilde{\psi}_j$ such that $\tilde{u}_j = \tilde{\psi}_{jy}$, and $\tilde{v}_j = -\tilde{\psi}_{jx}$. We use normal modes $(\tilde{\eta}, \tilde{\psi}_j, \tilde{p}_j, \tilde{\Gamma}) = [h, \phi_j(y), f_j(y), g] e^{i\alpha(x-ct)}$ where α is the wave number of the disturbance, g and h are constants, and $c = c_R + ic_I$ is the complex wave speed. The growth rate γ depends on the imaginary part of c only: $\gamma = \alpha c_I$. Linearizing the kinematic boundary condition yields $\tilde{\eta}_t(x, t) = -\tilde{\psi}_{x^*}(x, 0, t)$. Hence h is expressed in terms of the streamfunction: $h = \phi_1(0)/c$. The linearization of the horizontal and vertical components of the momentum equations (1) yields

$$m_j D (D^2 - \alpha^2) \phi_j - i \alpha f_j = i \alpha \frac{\text{Re}}{\text{Ca}} [(\bar{u}_j - c) D \phi_j - \phi_j D \bar{u}_j],$$

$$i \alpha m_j (D^2 - \alpha^2) \phi_j + D f_j = -\alpha^2 \frac{\text{Re}}{\text{Ca}} (\bar{u}_j - c) \phi_j,$$

where $m_1 = 1$, $m_2 = m$, $\text{Re} = \rho U_1 d_1 / \mu_1$ is the Reynolds number, and $\text{Ca} = \mu_1 U_1 / \sigma_0$ is the Capillary number. Eliminating the pressure disturbances f_j from these equations yields the well-known Orr–Sommerfeld equations for the streamfunctions:

$$m_j (D^2 - \alpha^2)^2 \phi_j = i \alpha \frac{\text{Re}}{\text{Ca}} [(\bar{u}_j - c) (D^2 - \alpha^2) \phi_j - \phi_j D^2 \bar{u}_j]. \quad (5)$$

The disturbance streamfunctions ϕ_j are subject to the boundary conditions at the plates and at the interface. The boundary conditions at the plates require

$$\phi_1(-1) = \phi_1'(-1) = \phi_2(n) = \phi_2'(n) = 0, \quad (6)$$

TABLE I. Parameters of normal modes (of wave number α) for $s \neq 0$ [$n > 1, m \neq n^2; \varphi = m + 3mn + 3n^2 + n^3, \psi = m^2 + 4mn + 6mn^2 + 4mn^3 + n^4$ and $\chi = (m-1)^2 n^2 (n+1)^2$].

Case			c	B_1	B_2
$n < \infty$	$m \neq 1$	Mode 1	$\frac{-i(n-1)M}{4(m-1)} \alpha$	$\frac{-4i(m-1)^2 s}{(n-1)(m-n^2)M} \alpha^{-1}$	$\frac{n^2}{m} B_1$
$n < \infty$	$m \neq 1$	Mode 2	$\frac{-2(m-1)(n+1)n^2 s}{\psi} - \frac{i(m-n^2)\varphi M}{4(m-1)\psi} \alpha$	$\frac{1}{2} \frac{m+3n^2+4n^3}{(n+1)n^2} - \frac{i\varphi\psi M}{16n^2\chi s} \alpha$	$-\frac{1}{2} \frac{4m+3mn+n^3}{mn(n+1)} - \frac{i\varphi\psi M}{16m\chi s} \alpha$
$n < \infty$	$m = 1$	Mode ₂ ¹	$\pm \frac{(1-i)}{2} \left(\frac{(n-1)n^2 Ms}{(n+1)^3} \right)^{1/2} \alpha^{1/2}$	$\frac{2(n^3+1)}{n(n^2-1)}$	B_1
$n = \infty$		Mode ₂ ¹	$\pm \frac{(1-i)}{2} (Ms)^{1/2} \alpha^{1/2}$	2	$-(1+i)(m-1) \left(\frac{s}{mM} \right)^{1/2} \alpha^{-1/2}$

where the symbol ' denotes differentiation with respect to y . Continuity of velocity at the interface yields

$$\phi_1(0) = \phi_2(0), \quad \phi_1'(0) - \phi_2'(0) = \frac{(1-m)}{m} \frac{s}{c} \phi_1(0). \quad (7)$$

After linearization, the normal stress condition, Eq. (2), yields

$$m\phi_2'''(0) - \phi_1'''(0) - 3\alpha^2[m\phi_2'(0) - \phi_1'(0)] = -i \frac{\alpha^3}{c} \phi_2(0). \quad (8)$$

The linearized tangential stress condition, Eq. (3), reads $m\phi_2''(0) - \phi_1''(0) + \alpha^2[m\phi_2(0) - \phi_1(0)] = iM\alpha g$, where $M = E\Gamma_0/\sigma_0$ is the Marangoni number. We replace the constant g in this equation by its expression from the linearized surfactant transport equation [derived from (4)], $\tilde{\Gamma}_t + \bar{u}_{1y}(0)\tilde{\eta}_x + \tilde{\psi}_{1xy}(x,0,t) = 0$, whence $g = (1/c)\phi_1'(0) + (s/c^2)\phi_1(0)$. As a result, the linearized tangential-stress balance condition is written purely in terms of streamfunctions:

$$m\phi_2''(0) - \phi_1''(0) + \alpha^2[m\phi_2(0) - \phi_1(0)] = iM \frac{\alpha}{c} \left[\phi_1'(0) + \frac{s}{c} \phi_1(0) \right]. \quad (9)$$

For each α , the linear (in ϕ_j) Eqs. (5)–(9) constitute an eigenvalue problem determining the (complex) phase velocity c .

In the limit of small wave numbers, and neglecting inertia terms, the Orr–Sommerfeld equations (5) reduce to

$$D^4 \phi_j = 0. \quad (10)$$

The general solutions are $\phi_j(y) = A_j + B_j y + C_j y^2 + D_j y^3$, where the constants A_j, B_j, C_j and D_j are determined, up to a normalization factor, by the boundary conditions. Adopting the normalization $A_1 = A_2 = 1$ and satisfying the four (plate) velocity conditions (6), leaves only B_1 and B_2 undetermined, with the streamfunctions expressed as

$$\phi_1(y) = 1 + B_1 y + (2B_1 - 3)y^2 + (B_1 - 2)y^3, \quad (11)$$

$$\phi_2(y) = 1 + B_2 y - \left(\frac{2}{n} B_2 + \frac{3}{n^2} \right) y^2 + \left(\frac{1}{n^2} B_2 + \frac{2}{n^3} \right) y^3. \quad (12)$$

Then Eqs. (7)–(9) are transformed (assuming $c \neq 0$) to the following system of three equations for B_1, B_2 , and c :

$$B_1 - B_2 = \frac{s}{c} \left(\frac{1}{m} - 1 \right), \quad (13)$$

$$6B_1 - \frac{6}{n^2} m B_2 - \frac{12m}{n^3} - 12 = \frac{i\alpha^3}{c}, \quad (14)$$

$$m \left(-\frac{4B_2}{n} - \frac{6}{n^2} \right) - 4B_1 + 6 = \frac{i\alpha M}{c} \left(B_1 + \frac{s}{c} \right), \quad (15)$$

where we have neglected terms which are higher than the leading-order terms by order α^2 . All results below follow from Eqs. (13)–(15). We will show that, for $m < n^2$, the two-fluid system is unstable if $s \neq 0$ (provided $M \neq 0$) and stable if $s = 0$. [This suggests to one that the last term of Eq. (15) is crucial for the destabilization.]

On using (13) to express B_2 in terms of B_1 , the quantity B_2 can be eliminated from both (14) and (15). Then B_1 is found in terms of c from (14) (assuming $m \neq n^2$). By substituting the resulting expression into Eq. (15), a single quadratic equation for the (phase velocity) c follows. Results for c, B_1 , and B_2 are given in Tables I and II for $s \neq 0$ and $s = 0$, respectively, and are most readily verified by direct substitution into the system (13)–(15) or, for $n = \infty$, into its analog described below. The equations have to be satisfied just to the leading order in all cases but the one with $s \neq 0, n < \infty, m \neq 1$, mode 2, in which cases the two leading orders are satisfied. We have also included in the tables the results for $n = \infty$ which will be discussed below.

For all the cases presented in the tables, there are two branches of solutions. Both modes are damped if $s = 0$. For $s \neq 0$, the first mode grows for $m < 1$ (and decays for $m > 1$), and the second one grows for $1 < m < n^2$ (and decays for $m < 1$ as well as for $m > n^2$); also, one of the modes is seen to

TABLE II. The same as Table I but for cases with $s = 0$.

Case		c	B_1	B_2
$n < \infty$	Mode 1	$-\frac{in^3}{12(m+n^3)} \alpha^3$	$\frac{n(m-n^2)}{2(m+n^3)M} \alpha^2$	B_1
$n < \infty$	Mode 2	$-\frac{in(m+n^3)M}{\psi} \alpha$	$-2 \frac{m+n^3}{n(m-n^2)}$	B_1
$n = \infty$	Mode 1	$-\frac{i}{12} \alpha^3$	$\frac{1}{12M} \alpha^4$	α
$n = \infty$	Mode 2	$-iM \alpha$	2	B_1

be growing if $m=1$ or/and $n=\infty$ (and the other mode is damped). Thus, for all $m < n^2$, there is always instability if $s \neq 0$, but the system is stable if $s=0$.

Note that the second mode for $s \neq 0$ and $m \neq 1$ has c real to the leading order in α . To find the leading-order growth rate αc_I for this mode, we write $c = c_0 + c_1 \alpha$ and $B_j = B_{j0} + B_{j1} \alpha$, and find the order α corrections, in particular c_I , as shown in Table I.

For the case $m=1$, $s \neq 0$, the two normal-mode waves, one growing and the other decaying, propagate in the opposite directions along the x -axis. The growth rate for $m=1$ changes as $\alpha^{3/2}$ while it changes as α^2 for the $m \neq 1$ cases. We conjecture that the nature of the singularity as $m \rightarrow 1$ is such that the range of α near $\alpha=0$ for which the $m \neq 1$ asymptotics is good, shrinks to zero in the limit. Singularities of this type are also encountered for the limits $s \rightarrow 0$ or $M \rightarrow \infty$. This issue cannot be resolved without going beyond the long-wave approximation. The pertinent work is in progress and the results will be published elsewhere.

Note that in the limit $n=\infty$, the eigenfunction (12) for the upper fluid no longer applies; instead, $\phi_2(y) = e^{-\alpha y}(1 + yB_2)$, which is the general (normalized) solution of $(D^2 - \alpha^2)^2 \phi_2 = 0$ satisfying the boundary condition $\phi_2(\infty) = 0$. Following the same procedure, one arrives at equations similar to (13)–(15). The changes are as follows: α is added to the right hand side of (13); terms containing n disappear from Eq. (14); and Eq. (15) becomes $-2B_1 - 2\alpha m B_2 + \alpha^2(2m-1) = (i\alpha M)/c(B_1 + s/c)$. This leads to the results given in the bottom lines of the tables, with the growth rate dependence on α being different from the case of a finite, no matter how large, n . [Thus, the long-wave results obtained for the semi-infinite layer are not necessarily a good approximation for any finite layer, even a very thick one.] One can see that the phase velocity is independent of the viscosity ratio m , and is proportional to $\alpha^{1/2}$, similarly to the finite n case with $m=1$.

The difference between the two disturbance modes can be illuminated by considering the ratio g/h (of the surfactant concentration amplitude to displacement amplitude) for the case of the plane Couette flow with $m=1$ and large n . One can see [since $h = \phi_1(0)/c = 1/c$ and $g = B_1/c + s/c^2 \approx s/c^2$] that $g/h \approx s/c \alpha \pm e^{i\pi/4}$, where the negative sign corresponds to the growing mode. Hence, for the growing mode, the surfactant concentration and the interface disturbance are out of phase by approximately $5\pi/4$, while for the decaying mode, the phase shift is $\pi/4$. For the growing mode, the surfactant concentration is a minimum, and thus the surface tension a maximum, approximately where the lower layer, the film, is thickest; and it is a maximum (surface tension minimum) where the film is thinnest. Thus there is a surface tension increase, and hence a flow, from troughs to peaks, which clearly causes the thickness contrast to grow. In contrast, for the stable mode, the spatial variations of the surfactant concentration and the interface are almost in phase, so there is now a fluid flow in the film from its peaks to troughs. This causes the disturbances to decay.

In conclusion, our investigation of the instability of a two-liquid planar system with a monolayer of insoluble sur-

factant on the interface reveals that some systems which are stable when stagnant can become unstable even in Stokes regimes of slow flow, provided there is a nonzero shear of velocity at the flat interface. Obviously, such instabilities can be overlooked unless nonstagnant cases are considered. In this respect, the instability uncovered in the present Letter is similar to the well-known Yih instability.² However, unlike the Yih instability, the present one does not depend on effects of inertia, and thus does not require invoking higher-order corrections to the Stokes-flow approximation. Also, we believe this to be the first example of a system which is stable without interfacial surfactant but unstable with the surfactant present.

For the future work, it would be interesting to extend the linear theory from small to all wave numbers. (This work is in progress; the results will be published elsewhere.) As regards the (weakly) nonlinear regimes, it is known⁴ that the shear flow can lead to saturation of instabilities. For the present instability, this would have an interesting twist that the same factor which facilitates the growth of infinitesimal disturbances at the early stages, later becomes a part of a nonlinear mechanism which works to hinder, and to eventually arrest, the growth of disturbances.

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