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# Instability of long-wavelength disturbances on gravity-modulated surfactant-covered thin liquid layers

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The effect of gravity modulation on long-wavelength disturbances at the free surface of a surfactant-covered thin liquid layer is analysed. The surfactants are assumed to be insoluble so that variations in their concentration along the free surface produce Marangoni flows in the underlying liquid. Lubrication theory is applied to obtain nonlinear partial differential equations that describe the behaviour of the free surface height and surfactant concentration, and the stability of these equations to small-amplitude disturbances is examined by applying Floquet theory. It is found that long-wavelength disturbances are destabilized by gravity modulation when surfactants are present, whereas such disturbances are stable when surfactants are absent. Results from additional calculations indicate that the instability becomes more difficult to excite as the Marangoni forces, body forces, capillary forces and surfactant diffusivity increase, and becomes easier to excite as the van der Waals forces increase.

## 1. Introduction

The adsorption of contaminants at the free surface of a liquid generally leads to a reduction in surface tension (Edwards, Brenner & Wasan 1991). If the concentration of the contaminants varies along the surface, surface tension variations will result and drive a flow in the underlying liquid. Known as Marangoni flows, these surface-tension-driven motions may be undesirable for applications such as coating (Scriven & Sternling 1964) and crystal growth (Carpenter & Homsy 1985). In microgravity environments, free surfaces in fluid systems are also subject to time-dependent gravitational forces. In the form of vertical vibration, such forces are capable of exciting standing waves at free surfaces, a topic well-studied since the seminal work of Faraday (1831). An issue of fundamental interest which arises from the above observations is the effect that gravity modulation has on disturbances at a contaminated free surface. Our goal is to address this issue for a class of flows where lubrication theory applies and the contaminants can be modelled as surfactants that are localized at the free surface.

Wave formation on vertically vibrated liquid layers has been analysed for inviscid (Benjamin & Ursell 1954; Miles & Henderson 1990), Newtonian (Kumar 1996; Perlin & Schultz 2000), and non-Newtonian liquids (Kumar 1999). In all cases, the vibration is assumed to be in the form of a sinusoidal modulation about a mean gravity. For viscous liquids, standing waves develop above a critical vibration amplitude that is a function of the vibration frequency. The standing waves correspond to time-periodic solutions of the linearized governing equations obtained through Floquet theory. A finite amplitude is required to excite the waves because the viscous dissipation of the liquid must be overcome. (As inviscid liquids lack a damping mechanism, a unique critical wavenumber is not predicted by theory for these liquids.) Measurements of the critical amplitude and wavenumber for both Newtonian (Bechhoefer *et al.* 1995) and non-Newtonian liquids (Raynal, Kumar & Fauve 1999; Wagner, Muller & Knorr 1999) are in excellent agreement with theoretical predictions.

The theoretical studies mentioned above are linear stability analyses and assume that the interface has a uniform surface tension. Recently, a linear stability analysis was performed by Kumar & Matar (2002) to account for surface tension variations due to the presence of an insoluble surfactant adsorbed at the interface. The analysis was valid for fluids of arbitrary viscosity and depth, and the effects of lateral boundaries were neglected. The main conclusion of Kumar & Matar (2002) is that surfactants can either raise or lower the critical vibration amplitude needed to excite the instability depending on the spatial phase shift between the surfactant concentration variations and surface deflections. If the concentration variations are in phase with the surface deflections (maximum concentration at wave crests), they will drive a Marangoni flow that pulls fluid away from the wave crests, and this will produce a larger critical amplitude. Similarly, if the concentration variations are out of phase with the surface deflections (minimum concentration at wave crests), they will drive a Marangoni flow that pulls fluid toward the wave crests, and this will produce a smaller critical amplitude. The above observations were found to be valid in the limit of zero surfactant diffusivity. For non-zero diffusivities, it was found that disturbances in the surface concentration of the surfactant simply decay exponentially on a time scale which is inversely proportional to the surface diffusivity of the surfactant. As a consequence, the time-periodic solutions of the linearized governing equations for this case correspond to standing waves for which the surface tension is spatially uniform, meaning that Marangoni flows are absent.

In this paper, we take a very different approach to the above problem by performing a lubrication analysis of the governing equations. We consider the thickness of the liquid layer to be much smaller than the wavelength of the interfacial disturbance, thereby allowing a separation of length scales that was not possible in Kumar & Matar (2002). With this approach, we obtain a coupled system of nonlinear partial differential equations describing the free surface height and the surfactant concentration as a function of time and the horizontal spatial coordinate. We find from these equations that the liquid layer is unstable to long-wavelength disturbances if it is covered by surfactants, while it is stable to such disturbances if the surfactants are absent. These results are valid for non-zero surfactant diffusivities, and represent standing wave solutions in which Marangoni flows are present. They can be contrasted to the results of Kumar & Matar (2002), who found that non-zero surfactant diffusivities prevented Marangoni flows from appearing in the standing wave solutions they constructed. The differences in the conclusions between that paper and the present one are due to the separation of length scales adopted here, which allows us to take advantage of certain asymptotic approximations.

## 2. Problem formulation

### 2.1. *Governing equations*

A thin layer of an incompressible Newtonian liquid, uniformly covered by a monolayer of insoluble surfactant, rests on a horizontal support. The support undergoes vertical sinusoidal oscillations of amplitude $a$ and frequency $\omega$. The vertical coordinate is denoted by $z$, such that the bottom of the plate is taken to be at $z = 0$, while the undisturbed free surface of the liquid is located at $z = h$. The horizontal and transverse coordinates are represented by $x$ and $y$, respectively. The effects of lateral boundaries are neglected.

The equations of momentum and mass conservation are respectively given by

$$\rho(\partial_t \boldsymbol{u} + \boldsymbol{u} \cdot \nabla \boldsymbol{u}) = -\nabla(p + \phi) + \eta_s \nabla^2 \boldsymbol{u} - \rho B(t) \boldsymbol{e}_z \quad \text{and} \quad \nabla \cdot \boldsymbol{u} = 0, \tag{2.1}$$

in which $\rho$ is the density, $\boldsymbol{u} = (u, v, w)$ represents the velocity, $t$ denotes time, $p$ is the pressure, $\eta_s$ is the liquid viscosity, and $\boldsymbol{e}_z$ is the unit vector in the $z$-direction. The variable $\phi = A/\zeta^3$ is a potential energy per unit volume term which represents van der Waals forces, where $A$ is the so-called Hamaker constant (Edwards *et al.* 1991) and $\zeta$ denotes the location of the free surface. The function $B(t) = g - a\cos(\omega t)$ is the modulated gravity wherein $g$ is the mean gravitational acceleration.

We now present the boundary conditions for these governing equations. The shear stress balance at the air–liquid interface (Deen 1998), located at $z = \zeta(x, y, t)$, is expressed by

$$\boldsymbol{t} \cdot \boldsymbol{\pi} \cdot \boldsymbol{n} = \boldsymbol{t} \cdot \nabla_s \sigma, \tag{2.2}$$

while the normal stress balance is given by

$$\boldsymbol{n} \cdot \boldsymbol{\pi} \cdot \boldsymbol{n} = -\sigma\kappa. \tag{2.3}$$

Here, $\sigma$ is the surface tension, $\kappa$ is the curvature, $\boldsymbol{t}$ and $\boldsymbol{n}$ are the tangent and normal unit vectors to the interface, and $\nabla_s = (\boldsymbol{\delta} - \boldsymbol{nn}) \cdot \nabla$ is the surface gradient operator. The tensor $\boldsymbol{\pi}$ is expressed by

$$\boldsymbol{\pi} = -(p + \phi - \rho B(t)\zeta)\boldsymbol{\delta} + \eta_s(\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T), \tag{2.4}$$

in which $\boldsymbol{\delta}$ is the identity tensor. The dynamics of the air overlying the liquid layer have been neglected in the stress balances. The kinematic boundary condition at $z = \zeta(x, y, t)$ is given by

$$(\partial_t + \boldsymbol{u}_s \cdot \nabla_s)\zeta = w_s, \tag{2.5}$$

where $\boldsymbol{u}_s$ and $w_s$ denote the velocity vector and its vertical component evaluated at $z = \zeta$. The equation of mass conservation of surfactant is given by

$$\partial_t \Gamma + \nabla_s \cdot (\boldsymbol{u}_s \Gamma) + (\boldsymbol{u}_s \cdot \boldsymbol{n})(\nabla_s \cdot \boldsymbol{n})\Gamma = \mathscr{D} \nabla_s^2 \Gamma, \tag{2.6}$$

where $\Gamma$ is the surfactant surface concentration and $\mathscr{D}$ is the surfactant surface diffusion coefficient (Stone 1990). In (2.6), the second and third terms on the left-hand side represent transport by convection and surface dilation, respectively, while the term on the right-hand side represents diffusive transport. Finally, no-slip and no-penetration conditions are applied at the support, located at $z = 0$:

$$u = w = 0. \tag{2.7}$$

### 2.2. *Scaling and asymptotic expansions*

In order to render these equations dimensionless, the following scaling is chosen:

$$(u, v) = (\omega h)(\tilde{u}, \tilde{v}), \quad w = (\epsilon \omega h)\tilde{w}, \quad (x, y) = L(\tilde{x}, \tilde{y}), \quad (z, \zeta) = h(\tilde{z}, \tilde{\zeta}), \tag{2.8}$$

$$t = (1/\omega)\tilde{t}, \quad p = (\eta_s \omega L/h)\tilde{p}, \tag{2.9}$$

where the tilde denotes dimensionless quantities. Here, we shall assume that the ratio of the film depth, $h$, to the wavelength of the interfacial disturbance, $L$, is $\epsilon = h/L \ll 1$.

In this limit, $k^*h \ll 1$, where $k^*$ is the dimensional disturbance wavenumber. For the surfactant concentration, $\Gamma$, and surface tension, $\sigma$, we choose the following scaling:

$$\Gamma = \Gamma_m \tilde{\Gamma} \quad \text{and} \quad \sigma = \sigma_m + \tilde{\sigma}\mathscr{S}, \tag{2.10}$$

where $\Gamma_m$ denotes the mean surfactant concentration and $\sigma_m$ is the surface tension corresponding to $\Gamma_m$. The spreading pressure is given by $\mathscr{S} = \sigma_0 - \sigma_m$, where $\sigma_0$ denotes the value of the surface tension for an uncontaminated liquid surface. We assume that the surfactant concentration is dilute enough to permit use of a linear equation of state:

$$\sigma = \sigma_0 + \left(\frac{\partial \sigma}{\partial \Gamma}\right)_{\Gamma=0} \Gamma. \tag{2.11}$$

Substitution of these scalings into the governing equations, boundary conditions and equation of state, yields a set of dimensionless equations to leading order in $\epsilon$. (The tilde notation is henceforth dropped.) For simplicity, we assume that the flow is two-dimensional. The $x$-component of the momentum conservation equation now becomes

$$Re(u_t) = -(p+\phi)_x + u_{zz} + O(\epsilon^2, \epsilon Re), \tag{2.12}$$

where $\phi = \tilde{A}/\zeta^3$, in which

$$\tilde{A} = \frac{A}{\eta_s \omega L h^2} \tag{2.13}$$

is a dimensionless Hamaker constant, and $Re$, the Reynolds number, is expressed by

$$Re \equiv \frac{\omega h^2}{\nu_s}, \tag{2.14}$$

in which $\nu_s$ is the kinematic viscosity of the liquid. The $z$-component of the momentum conservation equation is expressed by

$$0 = p_z + B(t) + O(\epsilon^2, \epsilon^2 Re), \tag{2.15}$$

in which $B(t) = \mathscr{B} - \mathscr{A}\cos(t)$ represents a modulated gravitational acceleration where

$$\mathscr{A} \equiv \frac{\epsilon \rho a h}{\eta_s \omega} \tag{2.16}$$

is the dimensionless amplitude of sinusoidal acceleration and

$$\mathscr{B} \equiv \frac{\epsilon \rho g h}{\eta_s \omega} \tag{2.17}$$

provides a measure of the significance of mean gravitational forces. The equation of continuity remains unaltered after the rescaling.

The shear stress balance becomes

$$u_z = \mathscr{M}\sigma_x + O(\epsilon^2), \tag{2.18}$$

in which $\mathscr{M}$ is a Marangoni parameter, which represents a ratio of Marangoni forces to viscous forces:

$$\mathscr{M} \equiv \frac{\epsilon \mathscr{S}}{\eta_s \omega h}. \tag{2.19}$$

The normal stress balance is expressed by

$$p = -\mathscr{C}\mathscr{M}\zeta_{xx} + O(\epsilon^2), \tag{2.20}$$

in which the parameter $\mathscr{C}$,

$$\mathscr{C} \equiv \frac{\epsilon^2 \sigma_m}{\mathscr{S}}, \tag{2.21}$$

is an inverse capillary number reflecting the importance of capillarity. Note that the potential effect of Marangoni stresses on capillarity has been neglected (see Appendix). The kinematic boundary condition at $z = \zeta$ is given by

$$\partial_t \zeta + \epsilon(u\zeta_x - w) = 0, \tag{2.22}$$

and the equation of surfactant mass conservation by

$$\partial_t \Gamma + \epsilon(u\Gamma)_x = \epsilon Pe^{-1}\Gamma_{xx}, \tag{2.23}$$

where $Pe$, which represents the ratio of surfactant transport by Marangoni stresses to that by surface diffusion, is expressed by

$$Pe \equiv \frac{\omega h L}{\mathscr{D}}. \tag{2.24}$$

Following scaling, the surfactant equation of state simply becomes $\sigma = 1 - \Gamma$. Finally, the no-slip and no-penetration conditions become $u = w = 0$.

We now demote the inertial terms in the equations of momentum conservation by setting $T = \epsilon t$, which yields

$$\epsilon Re(u_T + uu_x + wu_z) = -(p + \phi)_x + u_{zz} + O(\epsilon^2). \tag{2.25}$$

The kinematic boundary condition at $z = \zeta$ then becomes

$$\partial_T \zeta + u\zeta_x = w, \tag{2.26}$$

and the equation of surfactant mass conservation becomes

$$\partial_T \Gamma + (u\Gamma)_x = Pe^{-1}\Gamma_{xx}. \tag{2.27}$$

We define $\mathscr{R} \equiv \epsilon Re$, in which $Re \sim O(1)$, and expand all variables in powers of $\mathscr{R}$:

$$\left.\begin{array}{ll} u = u_0 + \mathscr{R}u_1 + \cdots, & w = w_0 + \mathscr{R}w_1 + \cdots, \\ p = p_0 + \mathscr{R}p_1 + \cdots, & \phi = \phi_0 + \mathscr{R}\phi_1 + \cdots, \\ \zeta = \zeta_0 + \mathscr{R}\zeta_1 + \cdots, & \Gamma = \Gamma_0 + \mathscr{R}\Gamma_1 + \cdots. \end{array}\right\} \tag{2.28}$$

Insertion of this expansion into the dimensionless governing equations yields the following equations to leading order in $\mathscr{R}$:

$$0 = -(p_0 + \phi_0)_x + u_{0zz}, \tag{2.29}$$

$$p_{0z} = -B(t), \tag{2.30}$$

$$u_{0z} = \mathscr{M}\sigma_{0x} \quad \text{on} \quad \zeta_0, \tag{2.31}$$

$$p_0 = -\mathscr{C}\mathscr{M}\zeta_{0xx} \quad \text{on} \quad \zeta_0, \tag{2.32}$$

$$\zeta_{0T} + u_0\zeta_{0x} = w_0 \quad \text{on} \quad \zeta_0, \tag{2.33}$$

$$\Gamma_{0T} + (u_0\Gamma_0)_x = Pe^{-1}\Gamma_{0xx} \quad \text{on} \quad \zeta_0. \tag{2.34}$$

The leading-order system of equations can be solved to yield the following:

$$u_0 = \mathscr{M}\sigma_{0x}z + \tfrac{1}{2}z(z - 2\zeta_0)(p_0 + \phi_0)_x, \tag{2.35}$$

$$w_0 = -\mathscr{M}\tfrac{1}{2}z^2\sigma_{0xx} + \tfrac{1}{2}z^2\zeta_{0x}(p_0 + \phi_0)_x - \tfrac{1}{6}(z^3 - 3z^2\zeta_0)(p_0 + \phi_0)_{xx}, \tag{2.36}$$

in which

$$p_0 = B(T)(\zeta_0 - z) - \mathscr{C}\mathscr{M}\zeta_{0xx}. \tag{2.37}$$

These can then be used to construct the leading-order evolution equations for $\zeta_0$ and $\Gamma_0$:

$$\zeta_{0T} = \left( \tfrac{1}{2}\mathscr{M}\zeta_0^2\Gamma_{0x} + \tfrac{1}{3}\zeta_0^3[B(t)\zeta_{0x} - \mathscr{C}\mathscr{M}\zeta_{0xxx}] - \tilde{A}\frac{\zeta_{0x}}{\zeta_0} \right)_x, \tag{2.38}$$

$$\Gamma_{0T} = \left( \mathscr{M}\zeta_0\Gamma_0\Gamma_{0x} + Pe^{-1}\Gamma_{0x} + \tfrac{1}{2}\Gamma_0\zeta_0^2[B(t)\zeta_{0x} - \mathscr{C}\mathscr{M}\zeta_{0xxx}] - \frac{3}{2}\tilde{A}\frac{\Gamma_0}{\zeta_0^2}\zeta_{0x} \right)_x, \tag{2.39}$$

where we have used $\sigma_0 = 1 - \Gamma_0$. Note that the separation of length scales ($h$ and $L$) adopted in this work has given rise to these nonlinear evolution equations, which is in contrast to the case of a layer of arbitrary depth (Kumar & Matar 2002) in which no such separation was possible.

For the case where surfactants are absent, we set $\Gamma_0 = 0$ in (2.38) and (2.39), which leaves us with an evolution equation for $\zeta_0$:

$$\zeta_{0T}^{(o)} = \left( \frac{\zeta_0^{(o)^3}}{3}[B^{(o)}(T)\zeta_{0x}^{(o)} - \tilde{\mathscr{C}}^{(o)}\zeta_{0xxx}^{(o)}] - \widetilde{A^{(o)}}\frac{\zeta_{0x}^{(o)}}{\zeta_0^{(o)}} \right)_x, \tag{2.40}$$

where the superscript ($o$) denotes the surfactant-free case; here $B^{(o)}(T) = \mathscr{B}^{(o)} - \mathscr{A}^{(o)}\cos(T)$. The scalings have also been changed: $\mathscr{S} \rightarrow \sigma_0$, and $\sigma_m \rightarrow \sigma_0$. That is, the dimensional surface tension has been scaled with the value of the surface tension associated with the clean interface, $\sigma_0$. Hence $\mathscr{M}$ and $\mathscr{C}$ now become $\mathscr{M} \rightarrow \mathscr{M}^{(o)} = \epsilon\sigma_0/\eta_s\omega h$ and $\mathscr{C} \rightarrow \mathscr{C}^{(o)} = \epsilon^2$. The new dimensionless group appearing in (2.40), $\tilde{\mathscr{C}}^{(o)}$, is now expressed by $\tilde{\mathscr{C}}^{(o)} = \mathscr{C}^{(o)}\mathscr{M}^{(o)} = \epsilon^3\sigma_0/\eta_s\omega h = \epsilon^3/Ca$, where $Ca$ is a capillary number; thus $\tilde{\mathscr{C}}^{(o)}$ is an inverse capillary number. The remaining dimensionless groups $\tilde{A}^{(o)}$, $\mathscr{A}^{(o)}$ and $\mathscr{B}^{(o)}$ remain unchanged from (2.13), (2.16) and (2.17), respectively.

It is, of course, possible to construct evolution equations for higher-order corrections to $\zeta$ and $\Gamma$. In the present work, we focus on the stability of the leading-order equations, which is discussed next.

## 3. Stability analysis

In this section, we linearize the lubrication equations developed above, (2.38)–(2.40), and examine their stability to small perturbations. Where surfactants are present, we linearize (2.38) and (2.39) about $\zeta_0 = 1$ and $\Gamma_0 = 1$ by introducing the expansion

$$(\zeta_0, \Gamma_0)(x,t) = 1 + (\hat{\zeta}(t), \hat{\Gamma}(t))\mathrm{e}^{\mathrm{i}kx} \tag{3.1}$$

into the lubrication equations, where $k$ is a dimensionless wavenumber. Where surfactants are absent, the surface deformation, $\zeta_0^{(o)}$, is expanded in a similar way. We begin by considering the latter case.

### 3.1. *Case I: Surfactants absent*

The linearized version of (2.40) is

$$\hat{\zeta}_T^{(o)} = \tfrac{1}{3}k^2\mathscr{A}^{(o)}\cos(T)\hat{\zeta}^{(o)} - \tfrac{1}{3}k^2(\mathscr{B}^{(o)} + \tilde{\mathscr{C}}^{(o)}k^2)\hat{\zeta}^{(o)} + \tilde{A}^{(o)}k^2\hat{\zeta}^{(o)}. \tag{3.2}$$

This equation can be integrated from 0 to $T$ to yield

$$\hat{\zeta}^{(o)} = \hat{\zeta}^{(o)}(T=0)\exp[\tfrac{1}{3}k^2\mathscr{A}^{(o)}\sin(T) - \tfrac{1}{3}k^2(\mathscr{B}^{(o)} + \tilde{\mathscr{C}}^{(o)}k^2)T + \tilde{A}^{(o)}k^2T]. \tag{3.3}$$

Equation (3.3) indicates that disturbances to the free surface will decay to zero as $T \to \infty$ provided that $\tilde{A}^{(o)} < [\mathscr{B}^{(o)} + \tilde{\mathscr{C}}^{(o)} k^2]/3$. This means that instability will not occur as long as forces due to van der Waals attraction are sufficiently weak compared to gravity and capillary forces.

According to (3.3), the vibration amplitude does not affect the long-time behaviour of the surface disturbances. Thus, in the asymptotic limit considered here ($\epsilon \ll 1$, $\mathscr{R} \ll 1$), vertical vibration will not lead to the generation of standing waves when surfactants are absent. This limit corresponds to long-wavelength disturbances, $k^* h \ll 1$. Disturbances which do not satisfy this requirement may still be excited by vertical vibration and lead to the formation of standing waves. Indeed, linear analyses of the Faraday instability indicate that vibration will always destabilize surface waves of a given critical wavelength.

Finally, we point out that the asymptotic analysis performed here is fundamentally different in nature to one recently reported in the literature (Cerda & Tirapegui 1998; Cerda, Rojas & Tirapegui 2000). In that analysis, the linearized Navier–Stokes equations and boundary conditions are used to develop an evolution equation for the interfacial disturbance. The equation is then solved to obtain a solution which is non-local in time, and the solution is expanded in terms of an infinite series of time derivatives. Truncation of the series leads to a Mathieu equation which does predict that vibration will destabilize surface disturbances. While the authors refer to their work as a 'lubrication approximation', it does not appear to be a traditional lubrication analysis like the one presented here. An additional difference is that the lubrication equations developed in the present manuscript could be used to study the nonlinear behaviour of long-wavelength disturbances and the effects of van der Waals attraction.

### 3.2. *Case II: Surfactants present*

The linearized version of (2.38) and (2.39) is

$$\hat{\zeta}_T = \tfrac{1}{3} k^2 \mathscr{A} \cos(T) \hat{\zeta} - \tfrac{1}{3} k^2 (\mathscr{B} + \mathscr{C} \mathscr{M} k^2) \hat{\zeta} - \tfrac{1}{2} k^2 \mathscr{M} \hat{\Gamma} + \tilde{A} k^2 \hat{\zeta}, \tag{3.4}$$

$$\hat{\Gamma}_T = \tfrac{1}{2} k^2 \mathscr{A} \cos(T) \hat{\zeta} - \tfrac{1}{2} k^2 (\mathscr{B} + \mathscr{C} \mathscr{M} k^2) \hat{\zeta} - k^2 (\mathscr{M} + Pe^{-1}) \hat{\Gamma} + \tfrac{3}{2} \tilde{A} k^2 \hat{\zeta}. \tag{3.5}$$

This is a system of first-order ordinary differential equations with time-dependent coefficients in which the free surface and surface concentration disturbances are coupled. In the absence of vibration, the coefficients are no longer time dependent and the stability of the equations can be analysed by applying the Routh–Hurwitz criterion (Coughanowr 1991). We have performed such an analysis, but simply note the result since the calculation is straightforward. The key finding is that disturbances are stable provided that van der Waals forces are sufficiently weak relative to gravity and capillary forces.

In the presence of vibration, time-periodic solutions are expected based on Floquet theory. To study these solutions, we follow a procedure similar to that used in linear analyses of the Faraday instability (Kumar & Tuckerman 1994). We begin by replacing $\hat{\zeta}(t)$ and $\hat{\Gamma}(t)$ by

$$\hat{\zeta}(t) = \mathrm{e}^{(s+\mathrm{i}\alpha)t} \sum_{n=-\infty}^{\infty} \zeta_n \mathrm{e}^{\mathrm{i}nt} \quad \text{and} \quad \hat{\Gamma}(t) = \mathrm{e}^{(s+\mathrm{i}\alpha)t} \sum_{n=-\infty}^{\infty} \Gamma_n \mathrm{e}^{\mathrm{i}nt}, \tag{3.6}$$

where $s$ represents the instability growth rate (real valued) and the value of $\alpha$ indicates whether the response of the standing waves to the modulation is subharmonic

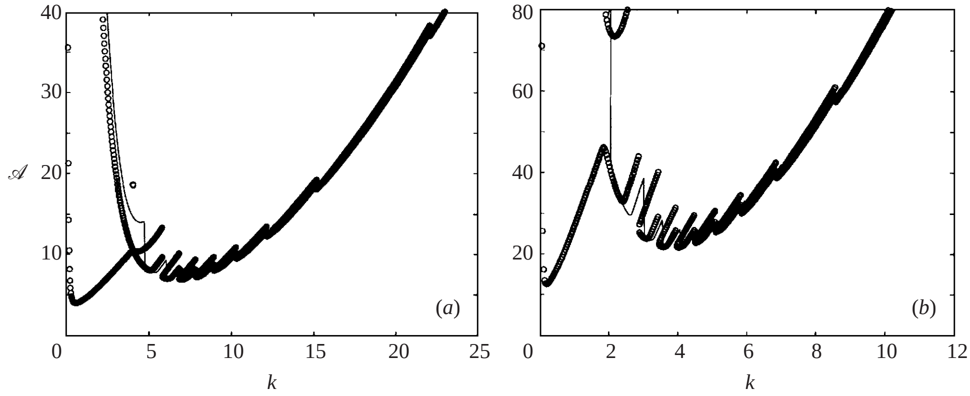

FIGURE 1. Neutral stability curves, $\mathscr{A}$ vs. $k$, for (a) $\mathscr{M} = 10$ and (b) $\mathscr{M} = 100$; the other parameters are $\mathscr{B} = 0.16$, $\mathscr{C} = 0.0075$, $Pe = 6.3 \times 10^5$, $\tilde{A} = 0$. The thin solid lines represent a subharmonic response and the circles represent a harmonic response. The curve for each type of response has a broad tongue-like shape that actually consists of a series of finer tongue-like curves. The harmonic curve has its global minimum closest to the $\mathscr{A}$-axis, meaning that the standing waves will respond harmonically to the vibration.

($\alpha = 1/2$) or harmonic ($\alpha = 0$). This then yields

$$(s + \mathrm{i}(\alpha + n))\zeta_n = \tfrac{1}{6}k^2\mathscr{A}(\zeta_{n+1} + \zeta_{n-1}) - \tfrac{1}{3}k^2(\mathscr{B} + \mathscr{C}\mathscr{M}k^2)\zeta_n - \tfrac{1}{2}k^2\mathscr{M}\Gamma_n + \tilde{A}k^2\zeta_n, \quad (3.7)$$

$$(s + \mathrm{i}(\alpha + n))\Gamma_n = \tfrac{1}{4}k^2\mathscr{A}(\zeta_{n+1} + \zeta_{n-1}) - \tfrac{1}{2}k^2(\mathscr{B} + \mathscr{C}\mathscr{M}k^2)\zeta_n \\ -k^2(\mathscr{M} + Pe^{-1})\Gamma_n + \tfrac{3}{2}\tilde{A}k^2\zeta_n. \quad (3.8)$$

It is possible to eliminate $\Gamma_n$ from (3.7) and (3.8) in order to arrive at the following recursion relation:

$$A_n\zeta_n = \mathscr{A}(\zeta_{n+1} + \zeta_{n-1}), \quad (3.9)$$

wherein

$$A_n = \frac{2(s + \mathrm{i}(\alpha + n) + \frac{1}{3}k^2D_n - \frac{1}{4}k^4\mathscr{M}D_n/C_n)}{\frac{1}{3}k^2 - \frac{1}{4}k^4\mathscr{M}/C_n}, \quad (3.10)$$

in which

$$C_n = s + \mathrm{i}(\alpha + n) + (\mathscr{M} + Pe^{-1})k^2,$$

$$D_n = \mathscr{B} - 3\tilde{A} + \mathscr{C}\mathscr{M}k^2.$$

The recursion relation (3.9) can be truncated at a finite value of $n$, say $n = N$, and converted into a matrix eigenvalue problem (Kumar & Tuckerman 1994). Solution of the eigenvalue problem for $s = 0$ produces a set of tongue-like neutral stability curves in the $(k, \mathscr{A})$-plane. The critical vibration amplitude needed to excite standing waves can be determined from the tongue tip closest to the $k$-axis, and this will also yield the critical wavenumber.

We have performed stability calculations using the recursion relation developed above, and figure 1 shows the results for two different values of $\mathscr{M}$. The other parameters have been fixed at typical values (see caption), and we take $N = 20$. For both $\mathscr{M} = 10$ and $\mathscr{M} = 100$, the tongue having the lowest amplitude corresponds to a harmonic response. The critical amplitude is larger for $\mathscr{M} = 100$, while the critical wavenumber is smaller. The increase in the critical amplitude can be explained by recognizing that when the free surface is perturbed, the surfactant concentration will be larger at the wave crests. This will produce a lower surface tension there than in the wave troughs, and liquid will flow from the crests to the troughs. As a result, it becomes more difficult to excite the instability. We have also performed additional calculations to study the effects of the other parameters on the instability behaviour. In these calculations, we fixed $\mathscr{M} = 10$ and varied one parameter at a time, holding the remaining ones at the values listed above. When $\mathscr{B} = 0.16$, the critical amplitude is larger than when $\mathscr{B} = 0$. Similarly, when $\mathscr{C} = 0.1$, the critical amplitude is larger than when $\mathscr{C} = 0.0075$. Since both gravity and surface tension tend to restore the system to an equilibrium state, increases in their mean values make it more difficult to excite the instability. When $Pe$ is decreased to unity, the critical amplitude increases compared to when $Pe = 6.3 \times 10^5$. This can be rationalized by recognizing that a lower value of $Pe$ corresponds to faster surfactant diffusion, and that the instability cannot be excited when the surfactant concentration is uniform. In all of the above cases, the critical wavenumber decreases whenever the critical amplitude increases. Finally, increasing $\tilde{A}$ from 0 to 0.05 decreases the critical amplitude and the critical wavenumber. As non-zero values of $\tilde{A}$ correspond to attractive van der Waals forces between the liquid and the plate, a smaller critical amplitude is required since the van der Waals forces will promote the growth of the surface waves.

## 4. Discussion and conclusions

In this work, we have addressed the issue of how gravity modulation affects disturbances at a contaminated free surface for a class of flows where lubrication theory applies and the contaminants can be modelled as insoluble surfactants. Our analysis led to nonlinear partial differential equations describing the behaviour of the free surface height and the surfactant concentration, and the stability of these equations to small perturbations was investigated. It was found that long-wavelength disturbances are destabilized when surfactants are present, in contrast to the case where surfactants are absent. Our results also indicate that the instability becomes more difficult to excite as the Marangoni forces, body forces, capillary forces and surfactant diffusivity become larger, but becomes easier to excite as the van der Waals forces increase.

The major contribution of this work is the presentation of a systematic lubrication analysis for vibration-driven standing waves. To our knowledge, such an analysis has not been reported in previous work, which also has not treated the effect of van der Waals forces. Our results also complement those of Kumar & Matar (2002), as they represent standing wave solutions valid for non-zero surfactant diffusivities that involve Marangoni forces in a non-trivial way. In addition, the nonlinear partial differential equations that we derive, (2.38)–(2.40), can be used to study the behaviour of large-amplitude disturbances, and a comprehensive numerical study will be reported in a separate work. The methods developed here may be useful in studying the behaviour of surface waves when both temperature gradients and gravity modulation are present. Then, Marangoni flows arise due to the temperature-dependence of the surface tension, and modelling of those flows is of considerable interest for microgravity applications (Birkh *et al.* 2001; Skarda 2001).

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## Appendix

Here, we illustrate the argument for neglecting the potential effect of Marangoni stresses on capillarity. Following the substitution of the scalings chosen in § 2.2 into the normal stress balance, the capillary term may be written as follows:

$$\frac{(\sigma_m + \tilde{\sigma}\mathscr{S})}{\eta_s L\omega}\epsilon^2\tilde{\zeta}_{xx} = \epsilon^2\mathscr{M}\left(\tilde{\sigma} + \frac{\sigma_m}{\mathscr{S}}\right)\tilde{\zeta}_{xx}, \tag{A 1}$$

where, for simplicity, we have only considered the streamwise direction. For dilute concentrations of surfactant such that $\mathscr{S}/\sigma_m \sim O(\epsilon^2)$ we can write the normal stress balance as

$$p = -\epsilon^2\frac{\sigma_m}{\mathscr{S}}\mathscr{M}\tilde{\zeta}_{xx} + O(\epsilon^2) = -\mathscr{C}\mathscr{M}\tilde{\zeta}_{xx} + O(\epsilon^2). \tag{A 2}$$

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