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Wind-generated waves in contaminated liquid films

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A uniform liquid film on a horizontal flat plate may be unstable to small disturbances when an air stream flows over the liquid surface. The stability of such films is examined for cases where the film is contaminated by an insoluble surface-active agent.

Two approximate analyses are given, which are applicable to liquid films at moderately large Reynolds numbers and at fairly small Reynolds numbers, respectively. These supplement previous work on uncontaminated films by Miles (1960), Cohen & Hanratty (1965) and Craik (1966).

At large liquid Reynolds numbers, the presence of surface contamination enhances stability due to increased dissipation in the viscous layer just within the liquid surface; but, at small liquid Reynolds numbers, there exists a class of disturbances for which surface elasticity may be destabilizing.

1. Introduction

Experiments concerning wind-generated waves in horizontal liquid films have been performed by Hanratty & Engen (1957), van Rossum (1959), Cohen & Hanratty (1965) and Craik (1966). It was observed by Craik that two distinct types of instability can occur, of which one is associated with very thin liquid films and the other with comparatively thick films. The three previous experiments concerned only the latter type of instability.

The interaction of the air flow and a small disturbance of the liquid surface gives rise to fluctuations of the normal and tangential stresses at the surface, and these are responsible for both types of instability. For very thin films, Craik has shown theoretically that a small periodic disturbance may be unstable under the joint action of the normal-stress component in phase with the wave displacement and the tangential-stress component in phase with the wave slope. However, for thicker films at moderately large liquid Reynolds numbers, instability is due primarily to the component of normal stress which is in phase with the wave slope. The theoretical stability problem for this case has been considered by Bondi (1942) and by Cohen & Hanratty.

The latter instability is due to the irreversible transfer of energy from the airflow to the disturbance, taking place through the non-conservative forces which act on the liquid surface. This instability occurs when the air flow is sufficiently large, for then the viscous dissipation within the liquid is insufficient to balance the energy transfer to a neutral wave. In contrast, the instability of very thin films is due to the action of conservative forces at the liquid surface, and it is virtually independent of the irreversible processes of viscous dissipation and of energy transfer to a neutral wave. A comprehensive discussion of such instability mechanisms is given by Benjamin (1964).

A further possible mechanism for instability has been proposed by Miles (1960). Due to the mean tangential stress exerted by the air flow, the primary velocity profile of the liquid varies linearly with the depth. At sufficiently large liquid Reynolds numbers, this flow may be unstable due to the action of viscous Reynolds stresses within the liquid, which transfer energy from the primary flow to the disturbance. In examining this mechanism, Miles neglected the stress fluctuations exerted by the airflow at the perturbed liquid surface.

It is well known that the presence of minute quantities of surface-active agent can greatly increase the rate of decay of waves in liquids of small viscosity, in the absence of an air flow. There have been several theoretical investigations of this phenomenon, notably by Levich (1962), Dorrestein (1951), van den Tempel & van de Riet (1965) and Miles (1967*a*). (A useful survey of the relevant work on this subject is given in the introduction to Miles's paper.) The results of these analyses are in substantial agreement with the experimental observations of Davies & Vose (1965) on the damping of capillary waves. A notable feature is that, as the amount of contamination is increased, the rate of decay is generally found to pass through a maximum for waves of a given frequency.

In the presence of an insoluble contaminant, the surface properties of the liquid may be identified with those of a visco-elastic membrane: for, deformation of the surface produces quasi-elastic forces proportional to strains and quasi-viscous forces proportional to temporal rates of strain. Such a mathematical model, incorporating both surface elasticity and surface viscosity, was used by Benjamin (1963) to examine the stability of a thin, contaminated liquid film flowing down an inclined plane. Here, Benjamin's representation is adopted to examine wind-generated waves in contaminated liquid films.

In the stability analysis, only two-dimensional harmonic disturbances need be considered; for, every periodic three-dimensional disturbance may be treated in terms of a corresponding two-dimensional problem (see Lin 1955, §§3.1, 5.2), and the development of relevant small initial disturbances may be deduced by Fourier synthesis. In addition to Squire's theorem (Squire 1933; Hanratty & Woodmansee 1965), the results of Watson (1960) and of Michael (1961) concerning the behaviour of three-dimensional, temporally-amplified disturbances are applicable to the present problem. However, care is required when dealing with the 'interfacial parameters'—for example, those denoting gravity and the surface properties of the film—in order that these results may retain their maximum physical significance.

For most of the analysis, the surface stresses which result from the interaction of the air flow and small periodic disturbances of the liquid surface are represented in a suitable parametric form: the substitution of actual estimates for these stresses is delayed until a fairly advanced stage. This approach has considerable advantages over that of Feldman (1957), whose treatment of the complete stability problem for a two-fluid system leads to great complexity. To evaluate the surface stresses, Cohen & Hanratty used a numerical technique developed by Miles (1962), which appears to yield satisfactory results when appropriate velocity profiles are adopted for the airflow. However, since the object of the present paper is to examine the role of surface contamination, it is here convenient to represent the surface stresses by less accurate, but simple, expressions which derive from the work of Benjamin (1959).

Since an analysis based on the exact solution of the equations of motion for the liquid would be extremely complicated, two separate approximate analyses are performed. The first of these is similar to the treatment of Cohen & Hanratty and involves the use of asymptotic approximations which are likely to be valid at moderately large values of the liquid Reynolds number. The second analysis is a straightforward extension of that given by Craik (1966), and is based on a method first introduced by Benjamin (1957). This analysis is applicable to thin films at fairly small Reynolds numbers.

2. Formulation of the problem

The initial formulation of the problem has much in common with that of Miles (1960) and of Craik (1966). For convenience, reference is made to the latter papers by prefixing the letters 'M' and 'I' respectively to the appropriate equation number. (The list of symbols given in I may be found useful by the reader.) The motion of the liquid is assumed to be laminar, and the response of the liquid to random turbulent fluctuations in the air flow is regarded as small (as was certainly the case in the experiments of Craik 1966). For channel flows, the air stream exerts a constant mean tangential stress on the liquid surface; also, the pressure gradient in the direction of motion produces negligible curvature of the liquid velocity profile whenever the depth of the air-phase is large compared with that of the liquid. We therefore take the mean velocity profile in the liquid to be linear. The experimental results shown in figure 4 of I confirm that this is a good approximation for the films under discussion. The present formulation is also adequate for airflows of boundary-layer type: for, even though the mean tangential stress exerted by the air stream, and also the mean film thickness, may vary with distance, the length scale associated with this variation is likely to be much larger than the wavelengths of all relevant disturbances.

All quantities are made dimensionless with respect to the film thickness h, the velocity V of the liquid surface and the liquid density ρ. The Reynolds number of the liquid film is defined as $R = Vh/\nu$ where ν is the kinematic viscosity of the liquid. The relationship connecting the primary motion of the liquid and that of the air is

$$\mu V/h = \mu_a U_i', \tag{2.1}$$

where $\mu (= \rho\nu)$ and $\mu_a (= \rho_a \nu_a)$ are the viscosity coefficients of the liquid and air respectively, ρ_a is the air density and U_i' is the dimensional velocity gradient of the air flow at the air-liquid interface. Dimensionless co-ordinates x and y are chosen such that the x-axis is parallel to the direction of flow and the y-axis is vertically upwards. In contrast to the notation of I, the rigid boundary is here denoted by $y = 0$ and the surface of the undisturbed film by $y = 1$. This agrees with Miles's notation and therefore facilitates comparison with his results. With this choice of co-ordinates, the dimensionless primary velocity profile in the liquid is

$$\bar{u}(y) = y. \tag{2.2}$$

A sketch of this configuration is shown in figure 1.

The vertical displacement of the liquid surface due to a small two-dimensional wave-like perturbation is represented by

$$y-1 = \eta(x,t) = \delta e^{i\alpha(x-ct)}. \tag{2.3a}$$

Here, α is the dimensionless wave-number, assumed to be real, and c is the dimensionless wave velocity, which may be complex with real and imaginary parts c_r and c_i. The associated horizontal displacement of particles comprising the liquid surface is denoted by

$$\xi(x,t) = \hat{X}\eta(x,t), \tag{2.3b}$$

where $\hat{X}$ is a complex quantity.

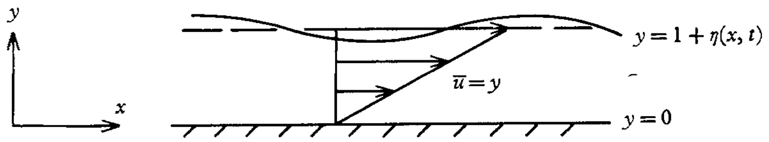

FIGURE 1. Sketch of shear flow and surface disturbance.

Because of continuity, a perturbation stream function may be introduced, of the form

$$\psi(x,y,t) = -\phi(y)\,\eta(x,t),$$

such that the perturbation velocity components are

$$u = \psi_y = -\phi'\eta, \quad v = -\psi_x = i\alpha\phi\eta, \tag{2.4a,b}$$

where the prime denotes differentiation with respect to y. Substitution of (2.4a,b) into the linearized equations of motion leads to the results

$$\phi^{\mathrm{iv}} - 2\alpha^2\phi'' + \alpha^4\phi = i\alpha R(y-c)\,(\phi''-\alpha^2\phi), \tag{2.5}$$

$$p = [(y-c)\,\phi' - \phi - (i\alpha R)^{-1}(\phi''' - \alpha^2\phi')]\,\eta(x,t), \tag{2.6}$$

$$\tau = R^{-1}(u_y + v_x) = -R^{-1}(\phi'' + \alpha^2\phi)\,\eta(x,t), \tag{2.7}$$

where p and τ are the perturbations of pressure and shear stress respectively.

The requirement that the perturbation velocity components should vanish at the wall yields the two boundary conditions

$$\phi(0) = \phi'(0) = 0. \tag{2.8a,b}$$

Also, the linearized kinematic surface conditions are

$$\frac{D\eta}{Dt} = v, \quad \frac{D\xi}{Dt} = u + \frac{d\bar{u}}{dy}\eta, \quad y = 1,$$

where the operator D/Dt denotes the time derivative following the motion of the undisturbed liquid surface. These lead to the results

$$\frac{\phi(1)}{1-c} = 1, \quad \hat{X} = \frac{i[\phi'(1)-1]}{\alpha(1-c)}. \tag{2.9a,b}$$

The remaining boundary conditions concern the normal and tangential stress perturbations at the air–liquid interface, and involve the stresses exerted by the air stream and by the liquid surface. As in I, the dimensionless normal and tangential stress perturbations exerted by the air stream at the interface are represented by

$$\sigma_{yy} = \Pi\eta(x,t), \quad \sigma_{xy} = \Sigma\eta(x,t). \tag{2.10a,b}$$

The parameters Π and Σ are complex and the subscripts r and i will be used to denote their real and imaginary parts.

As described in the introduction, the properties of a contaminated liquid surface are identified with those of a visco-elastic membrane. In its unstrained state, the surface is under a uniform tension γ equal to the mean surface tension; in dimensionless form, this tension is $T_0 = \gamma(\rho V^2 h)^{-1}$. Since the curvature after deformation is $O(\delta)$, the resulting elastic and viscous stresses make no first-order contribution to the normal stress, and the stress difference across the surface is simply T_0 times the curvature, as in the uncontaminated case. It follows that the linearized normal-stress relationship at the surface is identical to that given in I, namely,

$$-p + 2R^{-1}v_y = T_0\eta_{xx} + (\Pi - G)\eta \quad (y = 1).$$

Here, G equals gh/V^2, where g is the gravitational acceleration. On using results (2.4a,b), (2.6) and (2.9a), this relationship becomes [cf. I (4.9) and M(2.10c)]

$$\tilde{\omega} \equiv (1-c)\phi' - \phi - (i\alpha R)^{-1}(\phi''' - 3\alpha^2\phi') - (T_0\alpha^2 + G - \Pi)(1-c)^{-1}\phi = 0 \quad (y = 1). \tag{2.11}$$

The tangential stress just inside the liquid surface must equal the sum of the tangential stresses exerted by the contaminated surface and by the air stream. The stresses due to elasticity and viscosity of the surface are given by Benjamin (1963) in the form

$$\overline{\sigma}_{xy} = \left(T_1 + K\frac{D}{Dt}\right)\frac{\partial^2\xi}{\partial x^2} = -\,[T_1 + i\alpha(1-c)K]\,\alpha^2\hat{X}\eta, \tag{2.12}$$

$$T_1 = \gamma_1(\rho V^2 h)^{-1}, \quad K = \kappa(\rho V h^2)^{-1},$$

where γ_1 is the sum of the elastic moduli of surface dilatation and shear, and κ is the sum of the surface dilatation and shear viscosities. The boundary condition expressing continuity of tangential stress at the surface is therefore

$$\tau = \sigma_{xy} + \overline{\sigma}_{xy}, \quad y = 1;$$

or, on using results (2.7), (2.9a,b), (2.10b) and (2.12),

$$\phi'' + [\alpha^2 + R\Sigma(1-c)^{-1}]\phi - i\alpha R[T_1(1-c)^{-1} + i\alpha K][\phi' - (1-c)^{-1}\phi] = 0 \quad (y = 1). \tag{2.13}$$

The stability problem is now completely specified by the fourth-order Orr–Sommerfeld equation (2.5) and the four boundary conditions (2.8a,b), (2.11) and (2.13). The resulting characteristic-value relationship between c and α involves several representative parameters: R and G derive from the properties of the liquid film, Π and Σ from the airflow and T_0, T_1, K from the nature of the liquid surface. When both T_1 and K are zero, the above boundary conditions are equivalent to those given in I for a clean film.

3. The surface stresses

The perturbations in normal and tangential stress exerted by the air flow are represented by the complex dimensionless parameters Π and Σ. The real parts Π_r, Σ_r denote stress components in phase with the periodic surface displacement $\eta(x,t)$, while the imaginary parts Π_i, Σ_i derive from stress components which are in phase with the wave slope $\partial\eta/\partial x$.

Estimates of Π and Σ, which are based on the work of Benjamin (1959), are given in I (5.2) and I (5.3). These estimates were derived for mean air-velocity profiles of boundary-layer type, on the basis of a 'quasi-laminar' model for the air flow. Such a model neglects all turbulent fluctuations, and there is evidence to suggest that the contribution of these fluctuations may indeed be small under laboratory conditions; however, they are probably important for large-scale motions such as ocean waves. (A recent paper by Miles (1967*b*) contains a critical appraisal of the quasi-laminar model.)

Craik (1965) has shown that Benjamin's estimates of Π and Σ may also be applicable to turbulent air flow in a channel of finite height, provided the wavelengths of relevant disturbances are not large compared with the channel height. However, these estimates are based on several additional assumptions concerning the nature of the mean air flow, all of which do not appear to be satisfied in the existing experiments on wave generation in liquid films. It is therefore unlikely that the estimates will accurately represent all components of the stress perturbations for these experiments: the representation of Π_r may be fairly accurate, but the estimates of Π_i, Σ_r and Σ_i are probably only of the correct order of magnitude. A brief discussion of the range of validity of Benjamin's results is contained in I, and a fuller account is given by Craik (1965).

As mentioned in the introduction, Cohen & Hanratty appear to obtain satisfactory estimates of the surface stresses by employing Miles's (1962) numerical method. However, the present paper does not aim to provide accurate quantitative results for particular air velocity profiles; rather, its purpose is to elucidate the effect of surface contamination upon the stability of liquid films. For this purpose, it is sufficient to represent the surface stresses in a simple parametric form suggested by Benjamin's results. As in I (5.2), we represent the component of normal stress in phase with the wave slope by

$$\Pi_i = \frac{\alpha}{R}\left(2 - \frac{s}{c_f}\right). \tag{3.1}$$

Here, c_f is a friction coefficient, defined by the expression

$$\nu_a U_i' = c_f U_0^2,$$

where U_0 may be taken as the maximum velocity of the air flow. The quantity s is dependent on the properties of the air flow and of the surface displacement; but it may conveniently be regarded as fulfilling a role similar to the

Jeffreys 'sheltering coefficient' (Jeffreys 1925). Since the value of s/c_f is independent of the properties of the undisturbed liquid film, it is instructive to compare the behaviour of liquid films with various surface properties, when s/c_f takes constant values. Accordingly, s/c_f is treated as a constant parameter in the following analysis. In particular, the estimate of s given by Benjamin is not used here; consequently, the present treatment does not depend on the accuracy of this result. The term $2\alpha/R$ in Π_i derives from the mean shear stress exerted at the liquid surface. For, the parameter Π represents the direct stress acting vertically on the surface, while the mean shear stress R^{-1} acts tangentially on the *perturbed* liquid surface: a simple transformation shows that the latter contributes a first-order term of $2i\alpha/R$ times the surface displacement η to the direct stress in the y-direction.

For the normal stress component in phase with the wave displacement, we have, as in I (8.1*a*),

$$\Pi_r = \frac{\alpha I}{Rc_f}, \quad I = \int_1^H \left(\frac{U}{U_0}\right)^2 e^{-\alpha y}\,\alpha\,dy, \tag{3.2}$$

where the limits 1 and H of the integral I denote the boundaries of the airflow. Also, the estimate I (5.3) for the complex tangential stress parameter Σ is

$$\Sigma = \frac{2\beta I}{3^{\frac{1}{2}}c_f}\,e^{\frac{1}{3}i\pi}\alpha^3(\alpha R)^{-\frac{4}{3}}, \quad 2\beta/3^{\frac{1}{2}} = 1{\cdot}372\left(\frac{\nu_a}{\nu}\right)^{\frac{2}{3}}\left(\frac{\rho_a}{\rho}\right)^{\frac{1}{3}}. \tag{3.3}$$

In the following analysis, I/c_f is regarded as a constant parameter.

4. The eigenvalue equation for moderately thick films

Four linearly independent solutions of equation (2.5) were found by Hopf (1914), and details of these are given also by Feldman (1957) and Miles (1960). Two of these are

$$\phi_1 = \cosh \alpha y, \quad \phi_2 = \sinh \alpha y, \tag{4.1a,b}$$

which are usually called the 'inviscid solutions'. Since the remaining two solutions, the 'viscous solutions', are rather unwieldy, we shall use asymptotic approximations which are valid for large values of αR. The simplest such approximations are given by Lin (1955) as

$$\phi_{3,4} = \text{const}\,(y-c)^{-\frac{5}{4}}\exp\left[\mp \tfrac{2}{3}(i\alpha R)^{\frac{1}{2}}(y-c)^{\frac{3}{2}}\right]\{1+(\alpha R)^{-\frac{1}{2}}f(y)+\ldots\}, \tag{4.2a,b}$$

where $i^{\frac{1}{2}} = \exp(\frac{1}{4}i\pi)$ and, for nearly real values of c, $y-c = (c-y)\exp(-i\pi)$ when $y < c_r$. Both viscous solutions fluctuate rapidly: the amplitude of the fluctuations associated with ϕ_3 decreases exponentially with distance from the wall, while the amplitude of those associated with ϕ_4 increases exponentially.

Throughout the following analysis, we shall use the above asymptotic approximations to the viscous solutions ϕ_3 and ϕ_4. These are valid when

$$(\alpha R)^{\frac{1}{2}}|1-c|^{\frac{3}{2}} \gg 1, \quad (\alpha R)^{\frac{1}{2}}|c|^{\frac{3}{2}} \gg 1. \tag{4.3a,b}$$

The same approximations were used by Cohen & Hanratty and by Feldman, but Miles's analysis differs in this respect. To examine real values of c between 0 and 1, Miles did not impose the condition (4.3*b*): instead, he introduced asymptotic approximations which are uniformly valid with respect to c as αR becomes large—a refinement which is clearly necessary when c is very small. However, the present analysis primarily concerns values of c which exceed unity, and, for these, the inequality (4.3a) is the more stringent. The conditions under which this equality is satisfied are examined later.

The function $\phi(y)$ may be represented as a linear combination of the inviscid solutions $\phi_{1,2}$ and the above asymptotic viscous solutions $\phi_{3,4}$. Substitution for ϕ in the boundary conditions (2.8a,b), (2.11) and (2.13) then yields four linear equations for the coefficients of $\phi_{1,2,3,4}$. In order that these equations should be consistent, their determinant must vanish; and this condition gives the eigenvalue equation for c.

Following Miles, we may simplify the determinantal equation by neglecting $\phi_3(1)$, $\phi_4(0)$ and their derivatives. This is justified since, because of large exponential factors,

$$\left|\frac{\phi_4(1)}{\phi_4(0)}\right|, \quad \left|\frac{\phi_3(0)}{\phi_3(1)}\right| \gg 1$$

for nearly real values of c, both when $0 < c_r < 1$ and when $c_r > 1$. The resulting determinantal equation is

$$\begin{vmatrix} \phi_{10} & \phi_{20} & \phi_{30} & 0 \\ \phi'_{10} & \phi'_{20} & \phi'_{30} & 0 \\ \tilde{\omega}_{11} & \tilde{\omega}_{21} & 0 & \tilde{\omega}_{41} \\ f_{11} & f_{21} & 0 & f_{41} \end{vmatrix} = 0, \tag{4.4}$$

where

$$\tilde{\omega}_{i1} = (1-c)\,\phi'_i - \phi_i - (i\alpha R)^{-1}(\phi'''_i - 3\alpha^2\phi'_i) - g(1-c)^{-1}\phi_i \quad (y = 1),$$

$$f_{i1} = \phi''_i + (\alpha^2+S)\,\phi_i - Q\phi'_i \quad (y = 1),$$

and

$$g = T_0\alpha^2 + G - \Pi, \quad S = (R\Sigma + Q)(1-c)^{-1}, \quad Q = i\alpha R[T_1(1-c)^{-1} + i\alpha K].$$

Here, the first subscript identifies the solution and the second the point of evaluation, which is either $y = 0$ or $y = 1$. Substitution for ϕ_1 and ϕ_2 yields the result

$$\frac{\tilde{\omega}_{41}}{f_{41}} = \frac{\tilde{\omega}_{21}(\phi'_{30}/\phi_{30}) + \alpha\tilde{\omega}_{11}}{f_{21}(\phi'_{30}/\phi_{30}) + \alpha f_{11}}. \tag{4.5}$$

Also, from the asymptotic approximations (4.2a,b) for ϕ_3 and ϕ_4, we have the results

$$\frac{\tilde{\omega}_{41}}{f_{41}} = \frac{-g}{i\alpha R(1-c)^2 + S(1-c) - Q(i\alpha R)^{\frac{1}{2}}(1-c)^{\frac{3}{2}}} + O(R^{-\frac{3}{2}}), \tag{4.6}$$

$$\frac{\phi'_{30}}{\phi_{30}} = -(-i\alpha Rc)^{\frac{1}{2}} + \frac{5}{4c} + O[(\alpha R)^{-\frac{1}{2}}]. \tag{4.7}$$

On introducing

$$\theta = \frac{Q}{[i\alpha R(1-c)]^{\frac{1}{2}}} = \left(\frac{i\alpha R}{1-c}\right)^{\frac{1}{2}}[T_1(1-c)^{-1} + i\alpha K],$$

the results (4.5), (4.6) and (4.7) together lead to the equation

$$\alpha \coth \alpha(1-c)^2-(1-c)-g-\alpha(-i\alpha Rc)^{-\frac{1}{2}}[\alpha(1-c)^2-\coth \alpha(1-c+g)]$$
$$=(1-\theta)^{-1}\left\{\frac{iS}{\alpha R}[\alpha \coth \alpha(1-c)-1]+\frac{\theta g \alpha \coth \alpha}{[i\alpha R(1-c)]^{\frac{1}{2}}}+\frac{2i\alpha}{R(1-c)}[(1-c)^2\alpha \coth \alpha+g]\right.$$
$$\left.+\frac{5i}{4Rc^2}[\alpha(1-c)^2-\coth \alpha(1-c+g)]\right\}, \qquad (4.8)$$

where terms of order $O(R^{-\frac{3}{2}})$, $O(QR^{-\frac{3}{2}})$ and $O(SR^{-\frac{3}{2}})$ have been omitted.

5. The stability condition

In the following, the real and imaginary parts of c, g and Σ are denoted by the subscripts r and i. Also, c_r is taken to be positive, since it may be shown that all disturbances with negative c_r are stable. It is further assumed that $|c_i|$ is small compared with both $|c_r-1|$ and c_r.

It is convenient to introduce the parameters Ω and Ξ, defined as

$$\Omega=\left(\frac{\alpha R}{2|1-c_r|}\right)^{\frac{1}{2}}\frac{T_1}{|1-c_r|}, \quad \Xi=\left(\frac{\alpha R}{2|1-c_r|}\right)^{\frac{1}{2}}\alpha K.$$

The real and imaginary parts of θ are then

$$\theta_r=\Omega-\Xi, \quad \theta_i=\frac{1-c_r}{|1-c_r|}(\Omega+\Xi),$$

and we have the identity

$$|1-\theta|^2=1+2(\Xi-\Omega)+2(\Omega^2+\Xi^2).$$

The real part of equation (4.8) yields the result

$$\alpha \coth \alpha(1-c_r)^2-(1-c_r)-g_r=O\{(\alpha R)^{-\frac{1}{2}}, \quad \Omega(\alpha R)^{-\frac{1}{2}}, \quad |\Sigma|/\alpha|1-\theta|, Kc_i, c_i^2\}. \qquad (5.1)$$

Thus, to a first approximation, the dimensionless wave velocity c_r is unaffected by the presence of surface contamination. On making use of result (5.1), the imaginary part of equation (4.8) may be written as

$$(\alpha^{-1}Rc_i+2)\,[2\alpha \coth \alpha\,(c_r-1)+1]-\left(\frac{s}{c_f}-2\right)+\left(\frac{\alpha R}{2c_r}\right)^{\frac{1}{2}}\operatorname{cosech}^2\alpha(c_r-1)^2$$
$$+O\{c_i(\alpha R)^{\frac{1}{2}}, \quad c_iRT_1, \quad \Omega, \ \Xi, \ \operatorname{cosech}^2\alpha\}$$
$$=g_r\alpha^{-2}(c_r-1)^{-2}\,|1-\theta|^{-2}\{R[(1-\theta_r)\,\Sigma_r-\theta_i\Sigma_i]-(2\alpha R|c_r-1|)^{\frac{1}{2}}$$
$$\times[\alpha \coth \alpha(c_r-1)+1]\,[\Omega^2+\Xi+\Xi^2]\}. \qquad (5.2)$$

Since $(\alpha R)^{\frac{1}{2}}$ is large, all terms of $O(1)$ may be omitted from this equation except when Ω and Ξ are small. But, for uncontaminated films with $\Omega=\Xi=0$, such terms are important at large values of α, since $\operatorname{cosech}^2\alpha$ becomes very small. Terms which are $O(\Omega)$, $O(\Xi)$ or $O(\operatorname{cosech}^2\alpha)$ may always be neglected.

On setting Ω and Ξ equal to zero in equations (5.1) and (5.2), the results of Cohen & Hanratty for uncontaminated films are recovered. For such films, if s/c_f is greater than 2, the surface stresses represented by Π_i and Σ_r are both destabilizing when c_r is greater than unity, and both are stabilizing when c_r is less than unity. (Note that equation (5.1) requires that the coefficient of c_i in (5.2) is positive when $c_r > 1$ and negative when $0 < c_r < 1$.) In the former case, the surface stresses supply energy to a wave which travels downstream relative to the liquid surface. In the latter, the wave travels upstream relative to the liquid surface and must expend energy in opposing the surface stresses: the air flow therefore extracts energy from such waves, and the destabilizing mechanism is provided by the Reynolds stresses in the liquid layer close to the wall. The waves examined by Miles were of the latter type, while the analysis of Cohen & Hanratty concerned waves for which c_r was greater than unity. When c_r is negative, it may be shown that the surface stresses and the Reynolds stresses in the liquid are both stabilizing.

If s/c_f is less than 2, the role of the stress component Π_i is reversed. This occurs when the contribution to Π_i due to the mean tangential stress at the liquid surface is sufficiently large to outweigh the contribution of the direct stress which acts normally on the wave. However, the work of Cohen & Hanratty and the experiments of Craik and van Rossum confirm that the magnitude of s/c_f is typically $O(10)$ or greater at relevant wave-numbers: consequently, it is here assumed that s/c_f is greater than 2. The role of the stress component Π_i is unchanged by the presence of surface contamination: it is destabilizing when $c_r > 1$ and stabilizing when $0 < c_r < 1$.

When c_r is large compared with unity and terms in Σ_r and Π_i are neglected, the results for uncontaminated films agree with those of Bondi (1942), which were obtained by neglecting the primary motion of the liquid film. Bondi's wave velocity is simply the classical value for waves propagating in a liquid otherwise at rest.

Since the overall effect of surface contamination is not immediately apparent from (5.2), we consider in turn its influence on the roles of (i) the variable tangential stress Σ and (ii) the bulk viscosity R^{-1}, both when $0 < c_r < 1$ and when $c_r > 1$.

(i) In the absence of surface contamination, the term of equation (5.2) involving the tangential stress Σ is stabilizing when $0 < c_r < 1$ and destabilizing when $c_r > 1$. When $0 < c_r < 1$, this stabilizing role diminishes as the surface contamination is increased, and the net effect of the tangential stress components Σ_r and Σ_i actually becomes destabilizing if $[\Omega(1+r)+\Xi(r-1)] > 1$, where $r = \Sigma_i/\Sigma_r$. (According to result (3.3), r equals $3^{\frac{1}{2}}$.) When $c_r > 1$, the tangential stress components Σ_r and Σ_i are always destabilizing, and their influence is a maximum at some finite values of Ω and Ξ. For example, when r equals $3^{\frac{1}{2}}$ and Ξ is zero, the appropriate term of equation (5.2) has a maximum value at

$$\Omega = (2^{\frac{1}{2}}-1)/(3^{\frac{1}{2}}-1),$$

which is almost three times that when Ω is zero.

When Ω or Ξ is large compared with unity, the influence of the tangential stress Σ on both classes of disturbance is small. Also, the role of the tangential stress perturbations is negligible compared with that of the normal stress perturbations for *all* values of Ω and Ξ, if

$$\frac{s}{c_f}-2 \gg \frac{R g_r|\Sigma|}{\alpha^2(c_r-1)^2}. \tag{5.3}$$

With the estimate of Σ given in (3.3), this condition is satisfied when αR is sufficiently large.

(ii) The influence of surface contamination upon the role of the bulk viscosity is represented by the last term of equation (5.2). Since, from (5.1),

$$\alpha \coth \alpha(c_r - 1) + 1 = \mathscr{g}_r (c_r - 1)^{-1},$$

this term is stabilizing both when $0 < c_r < 1$ and when $c_r > 1$. The expression $(\Omega^2 + \Xi + \Xi^2)|1-\theta|^{-2}$ is a maximum with respect to Ω when Ω equals $1+\Xi$, and it has an absolute maximum value of 1 when $\Omega = 1$ and $\Xi = 0$. Also, if Ω is fixed and less than $\frac{1}{2}$, the expression has its maximum value when Ξ is infinite; while, if Ω is greater than $\frac{1}{2}$, its maximum value occurs when $\Xi = 0$. When either Ξ or Ω becomes large compared with unity, the value of $(\Omega^2 + \Xi + \Xi^2)|1-\theta|^{-2}$ approaches $\frac{1}{2}$.

Evidently, the contribution (ii) is most important. When condition (5.3) holds, the term in Σ may always be neglected. But, even when this condition is not satisfied, these terms may still be negligible if Ξ or Ω is large compared with unity. If, in fact, the tangential stress perturbation is negligible, the maximum degree of stabilization occurs when the surface viscosity is zero and when the surface elasticity is such that Ω equals unity.

6. The neutral case

As in I, we introduce the new parameters $\tilde{G}$ and $\tilde{T}$, defined as

$$\tilde{G} = R^2 G = gh^3/\nu^2, \quad \tilde{T} = R^2 T_0 = \gamma h/\rho\nu^2. \tag{6.1}$$

These remain constant for a liquid film of constant thickness, even though its surface velocity, and hence its Reynolds number, may vary. The use of $\tilde{G}$ and $\tilde{T}$ rather than G and T_0 allows a clearer physical interpretation of the results than was possible in Miles's work, where stability curves were obtained for several constant values of $T_0 + G\alpha^{-2}$. Although the critical conditions for the onset of instability in a given liquid film may be deduced from these curves, they lack the immediate physical significance of results pertaining to constant values of $\tilde{G}$ and $\tilde{T}$.

A further advantage of these parameters is apparent when three-dimensional disturbances are considered. With stability curves corresponding to constant values of G and T_0, the results of Squire (1933) and of Watson (1960)—see introduction—hold only for changes in R due to changes in *viscosity*, since G and T_0 depend on the length and velocity scales h and V. However, when stability curves are derived for constant values of $\tilde{G}$ and $\tilde{T}$, the results of Squire and of Watson hold for changes in R due to changes in *velocity*, with constant viscosity and film thickness. Also, since the film thickness h remains unchanged, the work of Michael (1961) is applicable.

Analogous to Miles's result (M 5.4), a necessary condition for the existence of neutrally stable disturbances with $0 < c_r < 1$ is

$$\alpha \coth \alpha - 1 > T_0 \alpha^2 + G - \Pi_r.$$

With prescribed values of I/c_f, $\tilde{G}$ and $\tilde{T}$, this condition may be expressed in the form $R > R_0(\alpha)$, where R_0 is a known function of α. Therefore, for neutral waves to exist with $0 < c_r < 1$ in a liquid film of given thickness, the Reynolds number R must exceed the minimum value of the function $R_0(\alpha)$.

For the uncontaminated case, Cohen & Hanratty have calculated curves of neutral stability for disturbances with $c_r > 1$. These calculations were based on more accurate representations of the surface stresses Π and Σ than those used here, and their results reveal that such disturbances usually become unstable at Reynolds numbers substantially smaller than the corresponding minimum values of $R_0(\alpha)$. It follows that disturbances for which $0 < c_r < 1$ are unlikely to play a significant part in determining the stability of clean liquid films: consequently, the instability mechanism examined by Miles (1960) is probably unimportant in this context. This conclusion is supported by the experiments of Cohen & Hanratty and of Craik. In these, waves with $c_r > 1$ occurred at moderately large liquid Reynolds numbers, but no disturbances with $c_r < 1$ were observed. (Disturbances with $c_r < 1$ were observed by Craik in very thin films, but the present theory is not applicable to these, since the Reynolds numbers were small; instead, the theory of I applies.)

Since surface contamination exerts a similar stabilizing influence on both types of disturbance—see (ii) above—it may be expected that, for contaminated films also, disturbances with $c_r > 1$ are less stable than those with $0 < c_r < 1$. Accordingly we now restrict attention to disturbances for which $c_r > 1$.

At this stage, it is convenient to derive the condition for onset of Kelvin–Helmholtz instability, which is required later. When R is sufficiently large, equation (4.8) simplifies to

$$\alpha \coth \alpha (1-c)^2 - (1-c) - g = 0;$$

and, if the stress component in phase with the wave slope is neglected, this quadratic in $1-c$ is found to have real or complex roots according as the quantity

$$G + T\alpha^2 - \Pi_r + (4\alpha)^{-1} \tanh \alpha$$

is positive or negative. The existence of complex roots signifies the occurrence of Kelvin–Helmholtz instability. The condition for onset of this instability may be written in the form

$$\tilde{G} + \tilde{T}\alpha^2 - \alpha R I c_f{}^{-1} + (4\alpha)^{-1} \tanh \alpha R^2 = 0, \qquad (6.2)$$

on using results (3.2) and (6.1).

For moderately thick uncontaminated films, the minimum value of R at which Kelvin–Helmholtz instability may arise is much larger than that at which instability occurs due to energy transfer through the normal stress component Π_i. However, it will be seen that the Kelvin–Helmholtz mechanism may be of greater significance for contaminated films.

On setting c_i equal to zero, curves of neutral stability may be obtained from equations (5.1) and (5.2). For simplicity, we assume that condition (5.3) is satisfied, and neglect terms in Σ_r and Σ_i, together with other small terms. With the typical values $s/c_f = 34$ and $I/c_f = 160$, neutral curves of R against α, for $c_r > 1$, were thus obtained for several pairs of values for $\tilde{G}$ and $\tilde{T}$ and for various surface properties. These surface properties were characterized by constant values of

$$[j] = \frac{2(\Omega^2+\Xi+\Xi^2)}{|1-\theta|^2},$$

the chosen values being $[j] = 0$, 0·5, 1 and 2. The first of these corresponds to a clean surface, the third to large contamination with either Ω or Ξ much greater than unity, and the last to maximum stabilization with $\Omega = 1$, $\Xi = 0$. For comparison, the corresponding curves for onset of Kelvin–Helmholtz instability were obtained from equation (6.2). The chosen values of $\tilde{G}$ and $\tilde{T}$ were appropriate for water films of various thicknesses, with $\nu = 10^{-2}\,\mathrm{cm^2\,sec^{-1}}$ and $\gamma/\rho = 73\,\mathrm{cm^3\,sec^{-2}}$. (The value of γ/ρ appropriate for clean water is retained here, although, in practice, this value is somewhat reduced by the presence of surface contamination. The effect upon stability of this reduction in mean surface tension is likely to be small compared with that due to surface elasticity and surface viscosity.) To facilitate their physical interpretation, the results are presented as curves of dimensional wave-number $k = \alpha/h$ against the mean dimensional shear stress τ_0. The latter is a property of the air flow alone, and is related to the Reynolds number of the film and the maximum air velocity U_0 by the expressions

$$\tau_0 = R\mu^2\rho^{-1}h^{-2} = \rho_a c_f U_0^2.$$

In figure 2 are shown the neutral curves for water films of thickness 1 cm, while table 1 shows the least values of τ_0, with corresponding values of k, which can sustain neutral waves in water films of three different thicknesses with various surface properties.

From these results, the critical value of τ_0 is seen to be greatly increased by the presence of surface contamination; and, since τ_0 equals $\rho_a c_f U_0^2$ where U_0 is the maximum velocity of the air flow, the critical air velocity is correspondingly increased. For water films of thickness 1 cm, the critical air velocity is over twice that for a clean surface when $[j] = 0{\cdot}5$, and more than five times that for a clean surface when contamination is large ($[j] = 1$). Also, with maximum stabilization ($[j] = 2$), the critical air velocity actually exceeds that for the onset of Kelvin–Helmholtz instability, the latter being 16 times larger than the critical velocity for a clean surface. For thinner films, the proportional increases in critical air velocity are rather less, but are still considerable: e.g. for a film of thickness 0·06 cm, the critical air velocity with large contamination in 86 % greater than with a clean surface.

At typical wave-numbers, Ω is $O(1)$ when the contaminated surface possesses only a small amount of elasticity. For example, with a water film of thickness 0·2 cm, when k is $O(10)\,\mathrm{cm^{-1}}$ and τ_0 is $O(1)$ dyne $\mathrm{cm^{-2}}$, Ω is likely to be $O(1)$ when the surface elasticity coefficient γ_1 is $O(1)$ dyne $\mathrm{cm^{-1}}$. With these same values of h, k and τ_0, Ξ is $O(1)$ when the surface viscosity κ is $O(10^{-2})\,\mathrm{g\,sec^{-1}}$.

Values of γ_1 which are $O(10)$ dyne $\mathrm{cm^{-1}}$ are common for contaminating monolayers of surface-active agents, but values of κ as large as $O(10^{-2})\,\mathrm{g\,sec^{-1}}$ are unusual. Thus, for most monolayers and at typical wave-numbers, the stabilizing effect of surface elasticity is large, while the effect of surface viscosity is likely to be negligible. However, if the surface is contaminated by certain proteins, for which κ is large, the role of surface viscosity can become significant. Also, even for typical monolayers, the role of surface viscosity may be important at very large wave-numbers.

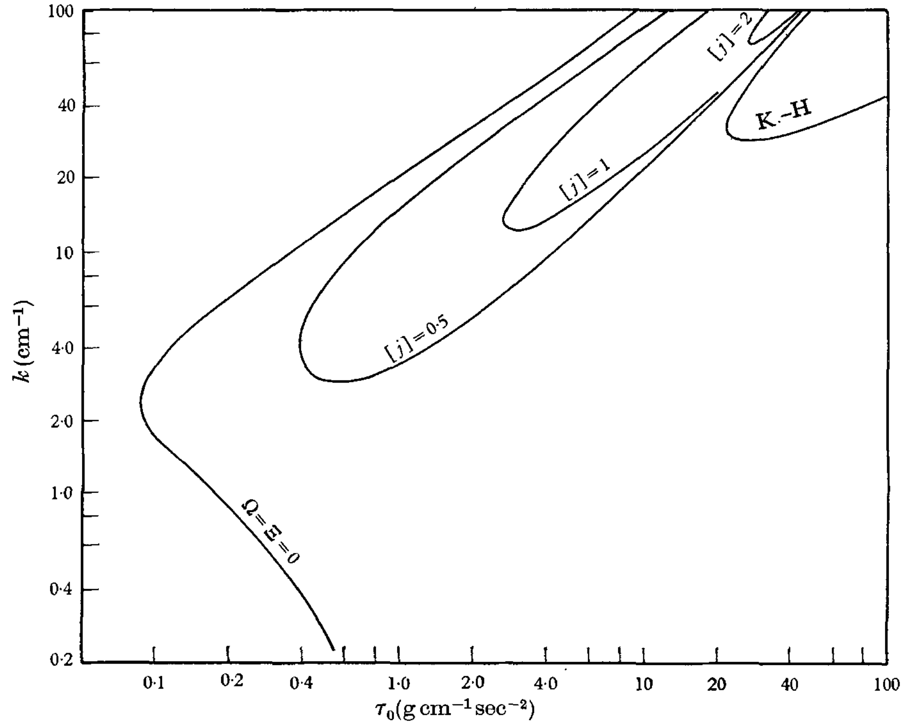

FIGURE 2. Neutral curves of wave-number k against mean shear stress τ_0 for water films of thickness 1 cm, with $s/c_f = 34$ and $I/c_f = 160$. The curves correspond, respectively, to a clean surface ($\Omega = \Xi = 0$), $[j] = 0{\cdot}5$, 1 and 2, and Kelvin–Helmholtz instability.

h(cm)	1·0		0·2		0·06	
	τ_0(g cm^{-1} sec^{-2})	k(cm^{-1})	τ_0	k	τ_0	k
Clean surface	0·084	2·4	0·38	7·0	1·40	4·5
$[j] = 0{\cdot}5$	0·39	4·3	0·717	8·0	2·22	15·0
Ω or $\Xi \gg 1$	2·70	13·5	2·75	13·8	4·86	22·0
$\Omega = 1, \Xi = 0$	27·0	75·0	27·0	75·0	27·0	75·0
Kelvin–Helmholtz *instability*	22·0	32·0	22·0	32·0	22·0	32·0

TABLE 1. Critical values of τ_0 and k for water films with various surface properties when $s/c_f = 34$ and $I/c_f = 160$

The observations of Keulegan (1951) and van Dorn (1953) indicate that the critical air velocity at which waves occur on relatively deep water may be increased by about an order of magnitude by the presence of surface contamination; while van Rossum (1959) found that the least air velocity capable of producing waves in a water film of thickness 0·06 cm may be increased from 7 to 10 msec^{-1} by the addition of surface-active agent. Unfortunately, no data are available concerning the elastic and viscous properties of the water surfaces in these experiments.

With $c_f = 2{\cdot}3 \times 10^{-3}$ and $\rho_a = 1{\cdot}29 \times 10^{-3}\,\mathrm{g\,cm^{-3}}$, the results shown in table 1 for a water film of thickness 0·06 cm yield critical air velocities of 690 cm sec^{-1} for a clean surface and 1240 cm sec^{-1} with large contamination. These values are consistent with van Rossum's observations; and the chosen values of s, I and c_f are believed to be appropriate for his experiments (see Craik 1965).

7. The approximations

If $\tilde{G}$ and $\tilde{T}$ are prescribed constants, some terms on the right-hand side of equation (5.1) may not be negligible compared with g_r when R is very large. These neglected terms are small compared with $G + T\alpha^2$ provided

$$\tilde{G} + \tilde{T}\alpha^2 \gg \alpha^{-1}(\alpha R)^{\frac{3}{2}}, \quad R^2 T_1. \qquad (7.1\,a, b)$$

Clearly, neutral curves obtained from the present approximations are likely to be accurate only if conditions (7.1) are satisfied. (Note that this difficulty does not arise when the prescribed constants are G and T instead of $\tilde{G}$ and $\tilde{T}$.) Fortunately, $\tilde{G} + \tilde{T}\alpha^2$ is usually large in cases of interest, and there is a considerable range of liquid Reynolds numbers for which conditions (4.3a, b) and (7.1) hold. Such Reynolds numbers may be termed 'moderately large'. With the estimate of Σ given in (3.3), condition (5.3) is likely to be met when (4.3a) is satisfied.

The accuracy of the asymptotic solutions (4.2a, b) depends on condition (4.3a) that $(\alpha R)^{\frac{1}{2}}|c-1|^{\frac{3}{2}} \gg 1$. This condition is usually well satisfied on the upper branches and near the minima of the neutral curves, provided the film thickness is not too small. However, it is *not* satisfied on the lower branches of the neutral curves at large R, since the wave velocity c tends to $1+$ there. In order to obtain accurate results for these branches, it would be necessary to introduce an asymptotic approximation to the viscous solution ϕ_4 which is uniformly valid with respect to $c-1$ as αR becomes large (cf. M 3.8). The neutral curves for the case of maximum stabilization are least accurate; for these, $(\alpha R)^{\frac{1}{2}}\,(c_r-1)^{\frac{3}{2}}$ is $O(1)$ in the vicinity of the critical values of τ_0. However, for the other neutral curves, conditions (4.3a) and (7.1) are satisfied near the critical values of τ_0.

8. The stability of thin contaminated films

We now consider the effect of surface contamination on wind-generated waves in thin liquid films. The method used is a straightforward extension of that described in I, the details of which need not be reiterated here. The stability problem is again characterized by the Orr–Sommerfeld equation (2.5) and the four boundary conditions (2.8a, b), (2.11) and (2.13); but, now, an approximate solution is sought under the assumptions

$$\alpha^2 \ll 1, \quad \alpha R < O(1), \quad \alpha R|c| < O(1). \qquad (8.1\,a, b, c)$$

It is readily shown that the governing equation, which expresses c in terms of α, R and the other parameters of the problem, is, to good approximation,

$$T_0\alpha^2+G-\Pi+\frac{3i\Sigma}{2\alpha} = (1-c)\left\{\frac{3}{i\alpha R}+\frac{6}{5}(1-c)-\frac{7}{8}+\frac{27}{5}\frac{\alpha^2}{i\alpha R}\right\}$$
$$-[T_1+i\alpha K(1-c)]\left\{\tfrac{3}{2}(c-1)^{-1}+3+\frac{i\alpha R}{5}(1-c)+\frac{2\alpha^2}{5}-\frac{i\alpha R}{4}\left(1+\frac{T_0\alpha^2+G-\Pi}{1-c}\right)\right\}. \tag{8.2}$$

When T_1 and K are both zero, this result is identical to equation I (6.5) which holds for uncontaminated films.

For neutrally stable disturbances, c is real and the real and imaginary parts of equation (8.2) are

$$T_0\alpha^2+G-\Pi_r-\tfrac{3}{2}(\Sigma_i/\alpha)+T_1(c-1)^{-1}\{\tfrac{3}{2}+\tfrac{1}{4}\alpha R\Pi_i+(c-1)(3+\tfrac{2}{5}\alpha^2)\}$$
$$=\tfrac{6}{5}(1-c)^2-\tfrac{7}{8}(1-c)+\alpha^2RK\{\tfrac{1}{5}(1-c)^2-\tfrac{1}{4}(1-c)-\tfrac{1}{4}(T_0\alpha^2+G-\Pi_r)\}, \tag{8.3}$$

$$-\Pi_i+\tfrac{3}{2}(\Sigma_r/\alpha)+(1-c)(\alpha R)^{-1}(3+\tfrac{27}{5}\alpha^2)+\alpha RT_1(1-c)^{-1}\{\tfrac{1}{5}(1-c)^2-\tfrac{1}{4}(1-c)$$
$$-\tfrac{1}{4}(T_0\alpha^2+G-\Pi_r)\}+\alpha K\{\tfrac{3}{2}+\tfrac{1}{4}\alpha R\Pi_i+(c-1)(3+\tfrac{2}{5}\alpha^2)\}=0. \tag{8.4}$$

The latter equation may be written as a quadratic in $(1-c)$; and, on invoking condition (8.1a) and making the additional assumptions

$$RK \leqslant O(1), \quad T_1\alpha^2R^2 \leqslant O(1), \quad R\Sigma_r \gg \alpha R|\Pi_i|, \quad \tfrac{1}{4}\alpha^2R^2T_1, \quad \alpha^2RK, \tag{8.5a,b,c}$$

approximate solutions are found to be

$$c-1=\tfrac{1}{4}R\Sigma_r[1\pm\sqrt{(1+\Lambda)}], \tag{8.6}$$

where
$$\Lambda=\tfrac{4}{3}T_1(\alpha/\Sigma_r)^2(T_0\alpha^2+G-\Pi_r).$$

Clearly, two types of neutral wave are possible, of which one travels upstream and the other downstream relative to the moving liquid surface. We now assume that

$$R\Sigma_r, \quad \Lambda^{\frac{1}{2}}R\Sigma_r \ll 1, \tag{8.7a,b}$$

in which case the wave velocity is close to unity for both types of disturbance.

By virtue of conditions (8.1), (8.5) and (8.7), terms in α^2, $\alpha R\Pi_i$ and α^2RK may be omitted from equation (8.3). Also, since c is close to unity, the terms $\tfrac{6}{5}(1-c)^2$ and $\tfrac{7}{8}(1-c)$ may also be neglected (cf. I, § 10). This leads to the simplified result

$$T_0\alpha^2+G-\Pi_r-\tfrac{3}{2}(\Sigma_i/\alpha)+T_1\{\tfrac{3}{2}(c-1)^{-1}+3\}=0. \tag{8.8}$$

Clearly, since $c-1$ is small, the term in $T_1\,(c-1)^{-1}$ is bound to play an important role, even when T_1 is itself small.

For amplified or damped disturbances, c is complex with real and imaginary parts c_r, c_i. For this case, results similar to (8.6) and (8.8) may be obtained, subject to the restriction that c_i is small. However, the resultant expressions for c_r and c_i are rather unwieldy, and it is more instructive to return to equation (8.2), keeping c complex but assuming that $|1-c|$ is small. Then, making simplifications similar to those above, we obtain the approximate equation

$$3(1-c)^2-i\alpha R\mathscr{G}(1-c)+i\alpha RT_1\{\tfrac{3}{2}+\tfrac{1}{4}i\alpha R(T_0\alpha^2+G-\Pi)\}=0, \tag{8.9}$$

where
$$\mathscr{G}=T_0\alpha^2+G-\Pi+\tfrac{3}{2}(i\Sigma/\alpha)+3T_1.$$

To the same degree of approximation as above, the imaginary part of Π may be disregarded.

When T_1 is zero, the relevant solution is $1-c = \frac{1}{3}i\alpha R\mathscr{G}$. (The solution $c = 1$ being extraneous in this case.) That is,

$$c_r - 1 = \tfrac{1}{2}R\Sigma_r - \tfrac{1}{3}\alpha R\Pi_i, \quad c_i = -\tfrac{1}{3}\alpha R[T_0\alpha^2 + G - \Pi_r - \tfrac{3}{2}(\Sigma_i/\alpha)],$$

in agreement with results I (7.2) and I(7.3). However, when T_1 is non-zero, both roots are permissible. Separating the real and imaginary parts of (8.9) and neglecting terms in Π_i and c_i^2, we obtain

$$\left.\begin{aligned} c_r - 1 &= \tfrac{1}{4}R\Sigma_r[1 \pm (1+\Lambda)^{\frac{1}{2}}] \pm \tfrac{2}{3}c_i\mathscr{G}_r(\alpha/\Sigma_r)(1+\Lambda)^{-\frac{1}{2}}, \\ \text{and} \qquad c_i[6(c_r-1) - \tfrac{3}{2}R\Sigma_r] &+ \alpha R\mathscr{G}_r(c_r-1) + \tfrac{3}{2}\alpha RT_1 = 0, \end{aligned}\right\} \tag{8.10}$$

where $\mathscr{G}_r$ denotes the real part of $\mathscr{G}$. On eliminating $(c_r - 1)$, these equations yield

$$\begin{aligned} c_i(R\Sigma_r)^{-1}(1+\Lambda)^{-\frac{1}{2}}\,[\tfrac{3}{2}(R\Sigma_r)^2(1+\Lambda) + \tfrac{2}{3}(\alpha R\mathscr{G}_r)^2] \\ + \tfrac{1}{4}\alpha R^2\mathscr{G}_r\Sigma_r[(1+\Lambda)^{\frac{1}{2}} \pm 1] \pm \tfrac{3}{2}\alpha RT_1 = 0, \end{aligned}$$

where the upper and lower signs correspond to those in (8.10). Since the coefficient of c_i is positive, the condition for instability is

$$R\mathscr{G}_r\Sigma_r[(1+\Lambda)^{\frac{1}{2}} \pm 1] \pm 6T_1 < 0,$$

which may be expressed more simply as

$$\mathscr{G}_r + \tfrac{3}{2}T_1(c_r - 1)^{-1} < 0, \tag{8.11}$$

where the value of $c_r - 1$ is now that given by equation (8.6).

Since $c_r - 1$ is a small *positive* quantity for the waves which are predicted to occur in uncontaminated films—we recall that Π_i is negative—the presence of even a minute amount of surface contamination might be sufficient to suppress these. However, there now exists a new class of disturbances, for which $c_r - 1$ is a small *negative* quantity, and for these the role of surface elasticity is strongly *destabilizing*. The appropriate condition for instability is then

$$GT_1(R\Sigma_r)^{-1}[(1+\Lambda)^{\frac{1}{2}} - 1]^{-1} > T_0\alpha^2 + G - \Pi_r - \tfrac{3}{2}(\Sigma_i/\alpha) + 3T_1. \tag{8.12}$$

As for waves in uncontaminated films, gravity and surface tension are stabilizing, and the normal stress component Π_r and the tangential stress component Σ_i are destabilizing. In addition, the term $3T_1$ on the right-hand side exerts a stabilizing influence, but this is small compared with the large destabilizing term on the left-hand side.

9. Discussion

In this analysis of thin films, the effect of surface viscosity has been shown to be negligible when $\alpha^2RK \ll 1$. Now, RK equals $\kappa/\mu h$ where κ is the dimensional surface-viscosity coefficient, and κ is of order $10^{-3}\,\mathrm{g\,sec^{-1}}$ or less for most monolayers. Taking $\kappa = 10^{-3}\,\mathrm{g\,sec^{-1}}$ and $\mu = 10^{-2}\,\mathrm{g\,cm^{-1}\,sec^{-1}}$ as typical values, it is seen that the inequality $\alpha^2RK \ll 1$ is satisfied for all relevant wave-numbers, provided h is less than about 0·1 cm. Therefore, for the thin films under investigation, the influence of surface viscosity is likely to be insignificant.

On the other hand, the presence of surface elasticity may have a marked effect, which is stabilizing or destabilizing according as the wave velocity is greater or less than unity. Now, T_1 is of the same order of magnitude as the restoring gravitational term G when $\gamma_1/\rho g h^2$ is $O(1)$; and, when the film-thickness h is $O(10^{-2})$ cm, this is so when γ_1 is $O(10^{-1})$ dyne cm^{-1}. Since T_1 is also multiplied by the large factor $(1-c)^{-1}$, it is clear that values of γ_1 as small as $O(10^{-2})$ dyne cm^{-1} may have a significant effect.It is relevant to note that values considerably larger than these are commonly found in tap water.

The main cause of this striking role of surface elasticity is a component

$$i\alpha T_1(1-c)^{-1}\eta$$

of the tangential stress $\overline{\sigma}_{xy}$ (see 2.9 and 2.12). This component derives from the fact that the mean velocity gradient $d\bar{u}/dy$ is non-zero at the liquid surface. Were this velocity gradient zero, as is the case for flow down an inclined plane, this term vanishes, leaving only that stress component in $\overline{\sigma}_{xy}$ which depends on the velocity perturbation u at the surface. It is the latter which gives rise to the term $3T_1$ on the right-hand side of result (8.12), and which is responsible for the stabilizing influence of surface elasticity in flow down an inclined plane (see Benjamin 1963).

When $1-c$ is real and positive, the stress component $i\alpha T_1(1-c)^{-1}\eta$ is in phase with the wave slope $\partial\eta/\partial x$, and therefore reinforces the tangential stress component $i\Sigma_i\eta$ due to the air flow: they drag liquid towards the crests and away from the troughs of a small wave-like disturbance. On the other hand, when $1-c$ is real and negative, the direction of the stress component $i\alpha T_1(1-c)^{-1}\eta$ is reversed and it acts in opposition to the destabilizing stress component $i\Sigma_i\eta$, thereby promoting stability. Gravity, surface tension and the direct stress component $\Pi_r\eta$ due to the air flow give rise to a periodic horizontal pressure gradient in phase with the wave slope, of magnitude $i\alpha(G+T_0\alpha^2-\Pi_r)\eta$. Provided $G+T_0\alpha^2-\Pi_r$ is positive, this pressure gradient tends to cause fluid beneath the crests to drain towards the troughs. The approximate condition for instability is then a simple kinematic one: namely, that there should be a net horizontal volume flux towards the crests and away from the troughs of some small wave-like perturbation.

The validity of the instability condition (8.12) is subject to the conditions (8.1a,b,c), (8.5a,b,c) and (8.7a,b). For the estimates of Σ and Π given in §3, it may be verified that all of these are generally satisfied for sufficiently thin films at relevant wave-numbers.

Since it appears that extremely small values of γ_1 should significantly affect the conditions governing instability in thin films, it is surprising that the theory for the uncontaminated case should agree so well with the experiments of Craik (1966). (The discrepancy between theory and experiment may be attributed to the approximate nature of the estimates of Σ_i and Π_r.) In these experiments, no rigorous precautions were taken to avoid surface contamination; therefore, one might expect that some small amount of surface elasticity was present. On the other hand, the present theory for the contaminated case does not seem to give results in agreement with the experimental observations. In particular, with the estimates of Σ and Π given in §3, the result (8.12) implies that disturbances of sufficiently large wavelength are always unstable, for all films to which the present theory is applicable. (However, this conclusion is rather sensitive to the dependence of Σ_r on α; and, in view of the approximate nature of the estimate (3.3), it should be treated with due scepticism.)

One may speculate that, since a new surface was continuously formed as the film flowed into the experimental apparatus, the contamination might have been much less than that commonly observed for surfaces which are exposed to the atmosphere for a longer time. This suggestion is supported by the experiments of van Dorn (1966) on wave damping in relatively deep water. In these, the effect of contamination was observed to increase with the age of the surface, reaching a maximum after about 1 h.

Another possibility deserves mention: since all visible waves in such thin films were non-linear—in the sense that their amplitude was comparable to the film thickness—and since c_r was 0·8 or less for these, the quantity $T_1(1-c_r)^{-1}$ is then much smaller than for truly infinitesimal waves. It is therefore conceivable that the influence of surface elasticity might be considerably diminished if the liquid film experienced *non-linear* initial disturbances: in view of the exceedingly thin film-thicknesses, it is not unlikely that such disturbances were present. However, these suggestions are purely speculative, and further progress must await a careful experimental investigation which incorporates precise measurements of the surface properties of the liquid films.

The role of surface contamination in moderately thick films, which was examined in the earlier part of this paper, is totally different from that just discussed. In such films, instability occurs when the viscous dissipation within the liquid is insufficient to balance the energy transfer from the mean air flow to a neutral wave. When $\alpha^{-1}R|c_r-1|$ is moderately large, wave energy is dissipated by viscosity in two fairly thin layers, one near the liquid surface and the other near the wall. For uncontaminated films, the dissipation in the surface layer is $O(\alpha R^{-1}|c_r-1|^{-1})$, but this may increase to $O(\alpha^{\frac{1}{2}}R^{-\frac{1}{2}}|c_r-1|^{-\frac{1}{2}})$ in the presence of surface contamination. By invoking boundary-layer approximations in such viscous layers, Miles (1967) has given a simple and illuminating account of this stabilizing role of surface contamination.

It at first seems surprising that, in figure 2, only the neutral curve corresponding to a clean surface should extend to small values of the wave-number k, while those for contaminated films reveal minimum values of k below which only damped disturbances may exist. This fact is due to the presence of a mean velocity gradient in the liquid: for, in (2.9*b*), a component of $\hat{X}$ derives from the velocity gradient at the liquid surface, and the resultant (stabilizing) component of the tangential stress $\overline{\sigma}_{xy}$—see (2.12)—becomes large at small values of $k(=\alpha/h)$ due to small values of $c-1$. Had this component been neglected, the remaining component of $\overline{\sigma}_{xy}$ would be found to exert a (diminished) stabilizing influence, but a branch of each neutral curve for contaminated films would extend to small values of k. It is only the latter component which is present when there is no air flow, and which is responsible for wave damping in liquids which are otherwise at rest.

Although the details of the present analysis differ from that of Miles due to the presence of a mean shear flow in the film, the nature of the stabilizing mechanism is essentially the same. In moderately-thick clean films, the flow perturbation near the surface is approximately irrotational; but when contamination is present, the motion possesses a significant rotational component due to the additional tangential stresses at the surface. The greatest dissipation occurs when this rotational component possesses a maximum share of the available wave-energy; and the analysis shows that this is so when $\Omega = 1$ and $\Xi = 0$. Thus, for such films, the forces of surface elasticity modify the oscillatory motion of the liquid so as to increase the rate of energy dissipation in the viscous layer near the surface.

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