The instability of liquid surfaces when accelerated in a direction perpendicular to their planes. I

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It is shown that, when two superposed fluids of different densities are accelerated in a direction perpendicular to their interface, this surface is stable or unstable according to whether the acceleration is directed from the heavier to the lighter fluid or vice versa. The relationship between the rate of development of the instability and the length of wave-like disturbances, the acceleration and the densities is found, and similar calculations are made for the case when a sheet of liquid of uniform depth is accelerated.

1. Introduction

If the horizontal surface of a liquid at rest under gravity is displaced into the form of regular small corrugations and then released, standing oscillatory waves are produced. Theoretically, a liquid could exist in a state of unstable equilibrium with flat lower horizontal surface supported by air pressure. If small wave-like corrugations were formed on this lower surface and then released, they might be expected to increase exponentially so long as their height was small compared with the wave-length.

This instability of the lower surface of a liquid might be expected to disappear if the liquid were allowed to fall freely, and to pass over into stability if the liquid were forced downwards with an acceleration greater than that of gravity. Similarly, the initial stability of the upper surface of a liquid might be expected to pass over into instability if the liquid were given a downward acceleration greater than that of gravity.

2. The instability of an interface between two fluids

Consider the interface between two fluids of densities ρ_1 and ρ_2. Assuming that they are accelerated vertically upwards with acceleration g_1, take horizontal and vertical axes which are fixed relative to the interface. The equations of motion relative to the accelerating axes are identical with those of a fluid at rest except that a term $-g_1\rho_1 y$ must be added to the pressure in the upper fluid and a term $-g_1\rho_2 y$ to that in the lower fluid. If both fluids are deep compared with the wave-length of the disturbance of the interface, the velocity potentials of the motion are

$$\phi_1 = A e^{-Ky+nt} \cos Kx \quad \text{in the upper fluid,} \tag{1}$$

$$\phi_2 = -A e^{Ky+nt} \cos Kx \quad \text{in the lower fluid.} \tag{2}$$

The equation of the surface of separation is then

$$\eta = A K n^{-1} e^{nt} \cos Kx. \tag{3}$$

The pressure in the upper fluid, neglecting squares of disturbance velocity, is

$$p_1 = p - (g+g_1)\rho_1 y + \rho_1 . \phi_1,$$

where p is the mean pressure at the interface and g is the acceleration of gravity.

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Similarly, the pressure in the lower fluid is

$$p_2 = p - (g + g_1)\rho_2 y + \rho_2 . \phi_2.$$

At the interface, therefore

$$-(g + g_1)(\rho_2 - \rho_1) y = (\rho_1 + \rho_2) n A \mathrm{e}^{nt} \cos Kx. \tag{4}$$

Hence from equation (3)

$$n^2 = -K(g + g_1)\frac{(\rho_2 - \rho_1)}{(\rho_2 + \rho_1)}. \tag{5}$$

If $(g + g_1)$ is negative, the acceleration downwards is greater than g and n^2 is positive. In this case the velocity potentials are of the form

$$\phi_1 = (A\mathrm{e}^{nt-Ky} + B\mathrm{e}^{-nt-Ky}) \cos Kx, \tag{6}$$

$$\phi_2 = -(A\mathrm{e}^{nt+Ky} + B\mathrm{e}^{-nt+Ky}) \cos Kx, \tag{7}$$

where n is taken to be the positive value of

$$\sqrt{\left(-K(g + g_1)\frac{(\rho_2 - \rho_1)}{(\rho_2 + \rho_1)}\right)}.$$

If the initial displacement of the interface from the plane $y = 0$ is

$$\eta_0 = C \cos Kx,$$

and the initial velocity is zero, equations (3), (6) and (7) become

$$\eta = C \cosh nt \cos Kx, \tag{8}$$

$$\phi_1 = \frac{nC}{K} \mathrm{e}^{-Ky} \sinh nt \cos Kx, \tag{9}$$

$$\phi_2 = -\frac{nC}{K} \mathrm{e}^{Ky} \sinh nt \cos Kx. \tag{10}$$

The initial disturbance of the interface will increase exponentially with nt until it has attained a magnitude which is no longer small in comparison with the wave-length. The rate of development of the instability is proportional to $\sqrt{\dfrac{(\rho_2 - \rho_1)}{(\rho_2 + \rho_1)}}$.

3. THE INSTABILITY OF A LIQUID SHEET OF FINITE THICKNESS h ACCELERATED BY AIR PRESSURE

In this case it will be assumed that the liquid sheet is of uniform thickness and is accelerated by air pressure acting on its upper surface. If the density of the liquid is great compared with that of air, the pressure may be assumed uniform over both the upper and lower surfaces.

Assuming in the first place that the elevations of the disturbances above their mean levels are of the form

$$\eta_U = \alpha \mathrm{e}^{nt} \cos Kx \quad \text{at the upper surface} \tag{11}$$

and $\qquad\qquad \eta_L = \beta e^{nt} \cos Kx$ at the lower surface, $\qquad\qquad$ (12)

these are consistent with a velocity potential

$$\phi = (A e^{Ky} + B e^{-Ky}) e^{nt} \cos Kx, \qquad\qquad (13)$$

provided that $\qquad\qquad \dfrac{\partial \eta_U}{\partial t} = -\dfrac{\partial \phi}{\partial y}$ at $y = 0$,

and $\qquad\qquad \dfrac{\partial \eta_L}{\partial t} = -\dfrac{\partial \phi}{\partial y}$ at $y = -h$,

so that $\qquad\qquad\qquad \alpha n = -K(A - B), \qquad\qquad (14)$

and $\qquad\qquad\qquad \beta n = -K(A e^{-Kh} + B e^{Kh}). \qquad\qquad (15)$

The pressure in the fluid (neglecting squares of disturbance velocity) is

$$p = P - \rho y(g + g_1) + \rho \dot{\phi},$$

where P is the pressure at the upper surface. The condition that p is constant at $y = \eta_U$ is $\qquad\qquad\qquad \alpha(g + g_1) = n(A + B), \qquad\qquad (16)$

while the condition at $y = \eta_L$ is

$$\beta(g + g_1) = n(A e^{-Kh} + B e^{Kh}). \qquad\qquad (17)$$

The four equations (14), (15), (16) and (17) can only be satisfied simultaneously if either $A = 0$ or $B = 0$.

If $B = 0$, then $\quad \alpha = \beta e^{Kh}, \quad \alpha n = -KA \quad$ and $\quad \alpha(g + g_1) = nA,$

so that $\qquad\qquad\qquad n^2 = -K(g + g_1). \qquad\qquad (18)$

If $A = 0$, then $\quad \alpha = \beta e^{-Kh}, \quad \alpha n = KB \quad$ and $\quad \alpha(g + g_1) = nB,$

so that in this case $\qquad\qquad n^2 = K(g + g_1). \qquad\qquad (19)$

By combining together solutions of these two types, the initial conditions at the top and bottom surfaces can be satisfied. As an example, take the case when the liquid is initially at rest but with the upper and lower surfaces corrugated into waves of amplitudes α_0 and β_0 respectively. Supposing that the liquid is accelerated downwards by applying a pressure at the upper surface, $(g + g_1)$ is then negative and the appropriate form for the velocity potential is

$$\phi = (A e^{Ky} \sinh nt + B e^{-Ky} \sin nt) \cos Kx, \qquad\qquad (20)$$

where n is the positive value of $\sqrt{\{-K(g + g_1)\}}$.

The elevation of the upper and lower surfaces are

$$\eta_U = \frac{K}{n}(-A \cosh nt - B \cos nt) \cos Kx, \qquad\qquad (21)$$

and $\qquad\qquad \eta_L = \frac{K}{n}(A e^{-Kh} \cosh nt - B e^{Kh} \cos nt) \cos Kx. \qquad\qquad (22)$

When $t = 0$, $\qquad\qquad \eta_U = \alpha_0 \cos Kx$ and $\eta_L = \beta_0 \cos Kx.$

Therefore
$$A + B = -\alpha_0 \frac{n}{K},$$
(23)

and
$$A e^{-Kh} + B e^{Kh} = -\beta_0 \frac{n}{K}.$$
(24)

Solving for A and B and substituting in equations (21) and (22), the elevations of the upper and lower surfaces become

$$\eta_U = \{(\alpha_0 - \beta_0 e^{-Kh}) \cosh nt - (\alpha_0 e^{-2Kh} - \beta_0 e^{-Kh}) \cos nt\} \frac{\cos Kx}{1 - e^{-2Kh}},$$
(25)

$$\eta_L = \{(\alpha_0 e^{-Kh} - \beta_0 e^{-2Kh}) \cosh nt - (\alpha_0 e^{-Kh} - \beta_0) \cos nt\} \frac{\cos Kx}{1 - e^{-2Kh}}.$$
(26)

In the experimental work described by Mr D. J. Lewis in part II (1950), various liquids were accelerated with an initial disturbance on the upper surface and with the lower surface initially flat. This corresponds to the case of $\beta_0 = 0$, when equations (25) and (26) reduce to

$$\eta_U = \alpha_0 \left\{ \frac{\cosh nt - e^{-2Kh} \cos nt}{1 - e^{-2Kh}} \right\} \cos Kx,$$
(27)

$$\eta_L = \alpha_0 \left\{ \frac{\cosh nt - \cos nt}{1 - e^{-2Kh}} \right\} e^{-Kh} \cos Kx.$$
(28)

The effect of the lower surface on the upper is represented by the terms in equation (27) containing e^{-2Kh} as a factor. If h is greater than $\frac{1}{3}\lambda$, where λ is the wave-length, e^{-2Kh} is less than 0·015. Neglecting the terms containing e^{-2Kh}, equation (27) becomes

$$\eta_U = \alpha_0 \cosh nt \cos Kx,$$
(29)

which is the form appropriate to a liquid of infinite depth.

In the experiments described in part II this approximation seems to be justifiable.

4. Conclusions

Defining the amplification factor of an unstable fluid surface as the ratio of the amplitude of the disturbance at any time to its initial value, we have for a vertical *downward* acceleration g_1

$$\frac{\eta}{\eta_0} = \cosh nt = \cosh \left\{ -K(g_1 - g) \frac{(\rho_2 - \rho_1)}{(\rho_2 + \rho_1)} \right\}^{\frac{1}{2}} t.$$
(30)

This applies to an initially disturbed interface between two fluids and also approximates closely to the behaviour of the upper surface of a sheet of liquid of density ρ_2 of thickness $h > \frac{1}{3}\lambda$ when accelerated by air pressure, in which case ρ_1 may be neglected by comparison with ρ_2. The acceleration is taken to begin at time $t = 0$, and at time t the liquid has moved downwards a distance s given by

$$s = \frac{1}{2} g_1 t^2,$$

so that
$$\frac{\eta}{\eta_0} = \cosh \sqrt{\left\{ \frac{4\pi s}{\lambda} \frac{(g_1 - g)}{g_1} \frac{(\rho_2 - \rho_1)}{(\rho_2 + \rho_1)} \right\}}.$$
(31)

If g_1 is large compared with g,

$$\frac{\eta}{\eta_0} = \cosh\sqrt{\left\{\frac{4\pi s}{\lambda}\frac{(\rho_2-\rho_1)}{(\rho_2+\rho_1)}\right\}} = \cosh\sqrt{\left\{4\pi m\frac{(\rho_2-\rho_1)}{(\rho_2+\rho_1)}\right\}}, \tag{32}$$

where m is the number of wave-lengths that the liquid has descended. Thus, when $g_1 \gg g$, the amplification of an unstable disturbance depends only on the ratio of the densities of the two fluids and the number of wave-lengths through which they have descended.

The experiments described in part II were designed to test the above theoretical conclusions and to find out how the unstable surface behaves when the amplitude of the disturbance becomes so great that the analysis based on the assumption that it is small ceases to be applicable.

REFERENCE

Lewis, D. J. 1950 *Proc. Roy. Soc.* A (in the Press).

The electronic structure of conjugated systems. VI

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This part reconsiders the previously neglected non-orthogonality of atomic orbitals. New generalized definitions of fractional bond order and charge distribution are introduced, and it is shown that under certain conditions, which are satisfied by most conjugated hydrocarbons, the overlap factor is completely irrelevant in calculating bond orders and charges. For heteromolecules this is not the case, and substantial changes may sometimes be made by its inclusion. The various polarizability coefficients introduced in earlier papers are always affected, but numerical results suggest that these changes are not very large.

INTRODUCTION

Previous papers in this series (Coulson & Longuet-Higgins 1947–8, referred to as I to V) have discussed in quite general terms the bond orders and charge distribution in conjugated systems; and have introduced various polarizability coefficients whose values showed how a molecule was likely to be affected by a change in the environment of any part of it. Applications were made to the law of alternating polarity in chemical reactions, to the vibrations of condensed molecules such as benzene and naphthalene, to the conjugating power of unsaturated free radicals, and to the differences between hydrocarbon systems such as naphthalene and heterosystems such as quinoline, in which a CH group has been replaced by a N atom. But in all this work it was explicitly supposed that all overlap integrals $S_{\mu\nu} = \int \phi_\mu \phi_\nu \, d\tau$ between distinct atomic orbitals ϕ_μ and ϕ_ν were identically zero. Such an approximation has almost universally been made hitherto, despite the fact that numerical calculations