# Finite-amplitude stability of a parallel flow with a free surface 

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The linearized problem of the instability of a layer of liquid flowing down an inclined plane was formulated by Yih (1954) and was solved by Benjamin (1957). It was found that the instability of such a film flow is initially due to long surface waves of infinitesimally small amplitudes. In the present study, a closed-form expression for the non-linear development of these long surface waves is obtained. It is shown that in the neighbourhood of the neutral curve an exponentially growing infinitesimal disturbance may develop into supercritically stable wave motion of small but finite amplitude if the surface tension of the liquid is sufficiently large. Theoretically obtained amplitudes of such waves are consistent with Kapitza's (1949) observation. The approach used in this analysis is a modification of the method used by Reynolds \& Potter (1967), who extended the method of Stuart (1960) and Watson (1960) in their study of the non-linear instability of plane Poiseuille and Poiseuille-Couette flow.

## 1. Introduction

Stuart (1960) has derived from the Navier-Stokes equation the following expression which governs the non-linear growth of a periodic finite disturbance in a parallel flow:

$$
\frac{1}{2} \frac{d A^{2}}{d t}=\alpha c_{i} A^{2}+a^{[2]} A^{4}+O\left(A^{6}\right)
$$

where $A$ is the disturbance amplitude, $\alpha$ the wave-number and $c_{i}$ the imaginary part of the eigenvalue obtained in the linearized stability theory. $a^{[2]}$ in the above equation, which was first suggested by Landau (1944), is sometimes called Landau's second coefficient. In the linearized theory, the last term in the above equation is negligibly small and the disturbance will grow or decay exponentially according to whether $c_{i}$ is positive or negative. However, the exponential growth of infinitesimal amplitude will immediately make the last term in the above equation significant and the sign of $a^{[2]}$ plays an important role in the nonlinear development of the initial disturbance. The analytic methods of determining the second Landau coefficient for plane Poiseuille flow have been given by Stuart himself and by Watson (1960) and also by Eckhaus (1965). The numerical evaluation of $a^{[2]}$ was carried out only recently by Reynolds \& Potter (1967) and also by Pekeris \& Shkoller (1967). By extending the method of Stuart
\& Watson, Reynolds \& Potter studied the instability of both plane Poiseuille flow and Poiseuille-Couette flow with respect to a finite-amplitude periodic oblique wave. Pekeris \& Shkoller assumed the periodic disturbance to be twodimensional and their method of analysis is primarily that of Eckhaus. The numerical result presented in these two papers indicates, as was pointed out by Reynolds, that plane Poiseuille flow will exhibit subcritical instability but no supercritical equilibrium flows. The non-linear instability of a boundary layer with respect to another type of three-dimensional disturbance is given by Benney (1961).

In the following sections, Stuart's approach is applied to investigate the stability of a falling liquid film with respect to a two-dimensional periodic disturbance of finite but small amplitude. Reynolds's expansion formalism is applied in formulating the boundary conditions. The second Landau coefficient and the wave speed in closed forms are then obtained by solving a sequence of non-homogeneous differential systems. Theoretical results are then compared with the observations of Kapitza (1949) and Binnie (1957).

## 2. Formulation of the problem

Consider a steady laminar layer of liquid flowing down an inclined plane under the action of gravity as shown in figure 1. A smooth surface of this film cannot be maintained at arbitrary Reynolds numbers. Two-dimensional periodic finite-


Fraure 1. Definition sketch.
amplitude wave motion has been observed by Kapitza (1949) and Binnie (1957) when the Reynolds number of the flow exceeds some critical values. In order to study the formation of such waves one chooses with Reynolds \& Potter (1967) the wave amplitude $A$, the phase $\theta$ of the wave motion and the distance measured normally downward from the unperturbed free surface as independent variables. In these independent variables, the Navier-Stokes equation can be written as

$$
\begin{gather*}
\frac{d A}{d t} \frac{\partial u}{\partial A}+\left\{\omega+\frac{d \omega}{d A}\left(t \frac{d A}{d t}\right)+\alpha u\right\} \frac{\partial u}{\partial \theta}+v \frac{\partial u}{\partial y}+\alpha \frac{\partial p}{\partial \theta}-\frac{\sin \beta}{F^{2}}-\frac{1}{R}\left\{\alpha^{2} \frac{\partial^{2} u}{\partial \theta^{2}}+\frac{\partial^{2} u}{\partial y^{2}}\right\}=0 \\
\frac{d A}{d t} \frac{\partial v}{\partial A}+\left\{\omega+\frac{d \omega}{d A}\left(t \frac{d A}{d t}\right)+\alpha u\right\} \frac{\partial v}{\partial \theta}+v \frac{\partial v}{\partial y}+\frac{\partial p}{\partial y}-\frac{\cos \beta}{F^{2}}-\frac{1}{R}\left\{\alpha^{2} \frac{\partial^{2} v}{\partial \theta^{2}}+\frac{\partial^{2} v}{\partial y^{2}}\right\}=0, \tag{1}
\end{gather*}
$$

where $R$ and $F$ are properly defined Reynolds and Froude numbers, $u$ and $v$ the velocity components in the $x$ - and $y$-directions, $\beta$ the angle of inclination of the plane, $\alpha$ the wave-number and $\omega$ the frequency of the wave motion assumed to depend on the amplitude $A . \alpha$ and $\omega$ are related to the phase $\theta$ of the basic wave by $\theta=\alpha x+\omega(A) t$.

The equation of continuity in the same independent variable is

$$
\begin{equation*}
\frac{\partial(\alpha u)}{\partial \theta}+\frac{\partial v}{\partial y}=0 \tag{2}
\end{equation*}
$$

which enables one to define a streamfunction $\psi$ such that

$$
\begin{equation*}
\alpha u=\partial \psi / \partial y, \quad-v=\partial \psi / \partial \theta \tag{3}
\end{equation*}
$$

In order to reduce (1) into a set of sequentially solvable equations, one formally introduces the following series expansions in terms of finite but small amplitudes:

$$
\begin{gather*}
\psi(A, y, \theta)=\alpha A^{n} \phi^{(k ; n)}(y) e^{i k \theta}+\alpha A^{n} \bar{\phi}^{(k ; n)}(y) e^{-i k \theta},  \tag{4}\\
A^{-1} \frac{d A}{d t}=a^{(0)}+A a^{(1)}+A^{2} a^{(2)}+\ldots=A^{n} a^{(n)},  \tag{5}\\
\omega+\frac{d \omega}{d A}\left(t \frac{d A}{d t}\right)=b^{(0)}+A b^{(1)}+\ldots=A^{n} b^{(n)}, \tag{6}
\end{gather*}
$$

where the bar denotes the complex conjugate. The superscript convention introduced in (4) indicates a sum over all $k$ and over all $n \geqslant k$. (For the details of this convention see Reynolds \& Potter (1967).) Since the unperturbed flow is the primary flow, one must have from (3) and (4)

$$
\begin{equation*}
\bar{u}=2 D \phi^{(0 ; 0)}=2 D \bar{\phi}^{(0 ; 0)}, \text { with } D \equiv d / d y \tag{7}
\end{equation*}
$$

Equation (5) is evidently valid in the neighbourhood of the neutral curve obtained with linearized theory and (6) is essentially a Poincaré eigenvalue stretching of the wave speed.

The pressure term in (1) can be eliminated by a cross-differentiation to yield a vorticity equation. Substituting (3) to (7) into the resulting vorticity equation and demanding the coefficient of $A^{n}$ in each harmonic to vanish, one has

$$
\begin{equation*}
L_{k n} \phi^{[k ; n]}=i \alpha c^{[n-1]} G \delta_{k 1}+H_{k n}, \tag{8}
\end{equation*}
$$

where $\delta_{i j}$ is the Kronecker delta,

$$
\begin{gather*}
i \alpha c^{(n)}=-a^{(n)}-i b^{(n)}, \\
G=\left(D^{2}-\alpha^{2}\right) \phi^{[i ; 1]}, \\
L_{k n}=i k\left[\left(-i(n / k) a^{(0)}+b^{(0)}+a \bar{u}\right)\left(D^{2}-k^{2} \alpha^{2}\right)-\alpha\left(D^{2} \bar{u}\right)\right]-R^{-1}\left(D^{2}-k^{2} \alpha^{2}\right)^{2},  \tag{9}\\
H_{k n}=-\left(m a^{[n-m]}+i k b b^{[n-m]}\right)\left(D^{2}-k^{2} \alpha^{2}\right) \phi^{(k ; m)}+F_{k n} /\left(1+\delta_{k o}\right),  \tag{10}\\
F_{k n} / \alpha=-\left(D \phi^{[k-j ; n-m]}\right)\left(i j\left[D^{2}-j^{2} \alpha^{2}\right] \phi^{[j ; m]}\right)-\left(D \bar{\phi}^{[j ; n-m]}\right) \\
\left.\times\left(i(k+j)\left[D^{2}-(k+j)^{2} \alpha^{2}\right] \phi^{[k+j ; m]}\right)-D \phi^{[k+j ; n-m]}\right)\left(-i j\left[D^{2}-j^{2} \alpha^{2}\right] \bar{\phi}^{[j ; m]}\right) \\
+\left(i(k-j) \phi^{[k-i ; n-m]}\right)\left(D\left[D^{2}-j^{2} \alpha^{2}\right] \phi^{[j ; m]}\right)+\left(-i j \overline{\left.\phi^{[j ; n-m]}\right)}\right. \\
\times\left(D\left[D^{2}-(k+j)^{2} \alpha^{2}\right] \phi^{[k+j ; m]}\right)+\left(i(k+j) \phi^{[k+j ; n-m]}\right)\left(D\left[D^{2}-j^{2} \alpha^{2}\right] \bar{\phi}^{[j ; m]}\right) . \tag{11}
\end{gather*}
$$

In the above expressions, the delimiter [ ] is used to indicate that the integer inside it must be greater than one and $\{k ; n\}$ means $0 \leqslant k \leqslant n \geqslant 1$. It will be seen shortly that ( 8 ) with proper boundary conditions is sequentially solvable. The decoupling of the original non-linear momentum equation is achieved by the expansion (4). In (4) it is implied in reality that the part of the solution of (1) which is of order $A^{n}$ depends only on the first ( $n-1$ ) harmonics (by the $n$th harmonics one means terms proportional to $\exp (i n \theta)$ ). It is this feature of the particular disturbance which enables one to build a non-linear solution on the solution obtained with the linearized theory.

The above recapitulation of Reynolds \& Potter's derivation of the vorticity equation which governs the amplitude variation function $\phi^{[k ; n]}$ for all harmonics is only for the purpose of providing sufficient information for further development. For the details of the derivation, readers are referred to Reynolds \& Potter's original paper. It should be pointed out that the expansion of the streamfunction (4) is different from Reynolds' only by a factor $\alpha$; this factor is introduced to avoid an overstretching of the shape of the free surface which appears in the boundary conditions. This modification also results in a difference by a factor $\alpha$ in the expression for $F_{k n}$.

The boundary condition at the fixed plate is the non-slip condition, i.e.

$$
\begin{equation*}
\phi^{[k ; n]}=D \phi^{[k ; n]}=0 \quad \text { at } \quad y=1 \tag{12}
\end{equation*}
$$

Neglecting the viscosity of the air and the inertia force at the interface, the free surface boundary conditions are simply the vanishing of tangential and normal forces, i.e.

$$
\begin{gather*}
\frac{\partial u}{\partial y}+\frac{\partial v}{\partial x}=0  \tag{13}\\
\left(-p+\frac{2}{R} \frac{\partial v}{\partial y}\right)+S k=0 \quad \text { at } \quad y=\eta(x, t) \tag{14}
\end{gather*}
$$

and
where $k$ is the curvature of the perturbed surface $y=\eta(x, t)$ and $S$ is a Weber number defined by $S=T / \rho d \bar{U}_{a}^{2}$ in which $T$ is the surface tension, and $\bar{U}_{a}$ is the average velocity of the primary flow and is given by

$$
\bar{U}_{a}=g d^{2} \sin \beta / 3 \nu
$$

in which $\nu$ is the kinematic viscosity of the fluid.
The pressure in the above boundary conditions is related to the velocity field by the momentum equation (1) and expressible in terms of $\phi^{[k ; n]}$. To do so one expands the pressure $p$ together with the streamfunction in terms of their harmonics, i.e.

$$
\begin{aligned}
& p=p^{(k)}(A, y) e^{i k \theta}+\bar{p}^{(k)}(A, y) e^{-i k \theta} \\
& \psi=\psi^{(k)}(A, y) e^{i k \theta}+\bar{\psi}^{(k)}(A, y) e^{-i k \theta} .
\end{aligned}
$$

Substituting the above expressions and (3) into the first equation of (1), one has
by virtue of (5) and (6) the following expression for the coefficient of $e^{i k \theta}$ :

$$
\begin{aligned}
\frac{d A}{d t} \frac{\partial D \psi^{(k)}}{\partial A} & +i k b^{(n)} A^{n} D \psi^{(k)}+\left[i m D \psi^{(k-m)} D \psi^{(m)}-i m D \psi^{(k+m)} D \bar{\psi}^{(m)}\right. \\
& \left.+i(k+m) D \bar{\psi}^{(m)} D \psi^{(k+m)}\right] /\left(1+\delta_{k o}\right) \\
& +\left[-i(k-m) \psi^{(k-m)} D^{2} \psi^{(m)}-i(k+m) \psi^{(k+m)} D^{2} \bar{\psi}^{(m)}\right. \\
& \left.+i m \bar{\psi}^{(m)} D^{2} \psi^{(k+m)}\right] /\left(1+\delta_{k o}\right)+i k \alpha^{2} p^{(k)} \\
& +\frac{\alpha^{2}}{R} k^{2} D \psi^{(k)}-\frac{1}{R} D^{3} \psi^{(k)}-\frac{\alpha \sin \beta}{2 F^{2}}=0
\end{aligned}
$$

The above equation is non-linear and coupled. In order to decouple it one seeks a solution of $p$ in the form $p^{(k)}=p^{(k ; n)} A^{n}$. Substituting this expression and (4) into the above equation and assuming the pressure distribution remains hydrostatic for the mean flow which is modified by the non-linear effect (i.e. $p^{(0 ; 0)}=\bar{p}^{(0 ; 0)}=y \cos \beta / 2 F^{2}$ and $p^{(0 ; n)}=\bar{p}^{(0 ; n)}=0$ for any $n>0$ ), one has from the coefficient of $A^{n}$ :

$$
\begin{align*}
-i k \alpha p^{[k ; n]}= & n a^{(0)} D \phi^{(k ; n)}+i k b^{(0)} D \phi^{(k ; n)} \\
& +\alpha\left[i m D \phi^{(m ; q)} D \phi^{(k-m ; n-q)}-i m D \bar{\phi}^{(m ; q)} D \phi^{(k+m ; n-q)}\right. \\
& +i(k+m) D \phi^{(k+m ; q)} D \bar{\phi}^{(m ; n-q)}-i(k-m) \phi^{(k-m ; q)} D^{2} \phi^{(m ; n-q)} \\
& \left.-i(k+m) \phi^{(k+m ; q)} D^{2} \bar{\phi}^{(m ; n-q)}+i m \bar{\phi}^{(m ; q)} D^{2} \phi^{(k+m ; n-q)}\right] \\
& +\frac{\alpha^{2}}{R} k^{2} D \phi^{[k ; n]}-\frac{1}{R} D^{3} \phi^{[k ; n]}+m a^{[n-m]} D \phi^{(k ; m)} \\
& +i k b^{[n-m]} D \phi^{(k ; m)} \text { for } n>0  \tag{15}\\
& \quad \frac{1}{\bar{R}} D^{3} \phi^{(0 ; 0)}-\frac{\sin \beta}{2 F^{2}}=0 \quad \text { for } \quad n=0 .
\end{align*}
$$

and
Since $D^{3} \phi^{(0 ; 0)}=D^{3} \bar{\phi}^{(0 ; 0)}=\frac{1}{2} D^{2} \bar{u}$ by virtue of (7), the above equation gives

$$
3 F^{2}=R \sin \beta
$$

which is the relation between the Froude number and the Reynolds number in the primary flow (Yih 1963).

On the other hand, the kinematic boundary condition at the free surface demands

$$
v=\frac{D \eta}{D t} \quad \text { at } \quad y=\eta(x, t)
$$

Expanding $v$ into Taylor's series around $y=0$, the above equation can be written as

$$
v(0)+v^{\prime}(0) \eta+\frac{v^{\prime \prime}(0)}{2} \eta^{2}+\ldots=\frac{\partial \eta}{\partial t}+\left[u(0)+u^{\prime}(0) \eta+\frac{u^{\prime \prime}(0)}{2} \eta^{2}+\ldots\right] \frac{\partial \eta}{\partial x}
$$

where primes denote differentiation with respect to $y$. With $u, v$ given by (3) and (4) one naturally seeks a solution of $\eta$ in the following form:

$$
\begin{equation*}
\eta=\eta^{(k ; n)}(x, t) A^{n} e^{i k \theta}+\bar{\eta}^{(k ; n)}(x, t) A^{n} e^{-i k t} . \tag{16}
\end{equation*}
$$

With exactly the same method which is used for obtaining (15), one can obtain from the above equation a lengthy recurrence formula for $\eta^{[k ; n]}$ which is given
by (Al) in the appendix. Thus, $p$ and $\eta$ can be expressed sequentially in terms of $\phi^{[k ; n]}$ and $\phi^{[k ; n]}(0)$, and thus the boundary conditions (13) and (14) can be reduced to expressions involving $\phi^{[k ; n]}(0)$ only. To see this, one expands (13) and (14) into Taylor's series around $y=0$, i.e.

$$
\begin{gathered}
\frac{\partial}{\partial y} \sum_{n=0}^{\infty}\left(D^{n} u / n!\right)_{o} y^{n}+\frac{\partial}{\partial x} \sum_{n=0}^{\infty}\left(D^{n} v / n!\right)_{o} y^{n}=0 \\
-\sum_{n=0}^{\infty}\left(D^{n} p / n!\right)_{o} y^{n}+\frac{2}{R} \frac{\partial}{\partial y} \Sigma\left(D^{n} v / n!\right)_{o} y^{n}+S \frac{\partial^{2} \eta}{\partial x^{2}}\left[1-\frac{3}{2}\left(\frac{\partial \eta}{\partial x}\right)^{2}\right]=0 \text { at } y=\eta
\end{gathered}
$$

Substituting (3), (4), (16) and $p=p^{[k ; n]} A^{n} e^{i k \theta}+\bar{p}^{[k ; n]} A^{n} e^{-i k \theta}$ into the above equation, one has from the coefficient of $A^{n}$ of each harmonic the boundary conditions (A 2) and (A3) given in the appendix with $p^{[k ; ~ n]}(0)$ given by (15) and $\eta^{[k ; n]}$ given by (Al).

Thus the problem is reduced to solving sequentially a set of non-homogeneous fourth-order systems of the following form:

$$
\begin{equation*}
L_{k n} \phi^{[k ; n]}=i \alpha c^{[n-1]} G \delta_{k 1}+H_{k n}, \quad U\left(\phi^{[k ; n]}\right)_{m}=\gamma_{m} \quad(m=1,2,3,4) \tag{17}
\end{equation*}
$$

in which $i \alpha c^{[n-1]}=-a^{[n-1]}-i b^{[n-1]}$ and the second equation stands for the four boundary conditions. (Two of them are always homogeneous but the two at the free surface may be non-homogeneous.) The solution of the above system is, in general, unique unless the associated homogeneous system

$$
L_{k n} \phi^{[k ; n]}=0, \quad U\left(\phi^{[k ; n]}\right)=0
$$

possesses eigensolutions. In that case the solution of the non-homogeneous system can be found only for particular values of $c^{[n-1]}$. One method of determining $d^{[n-1]}$ is to consider the adjoint homogeneous system (Ince 1926, chapter 9)

$$
\begin{equation*}
L^{*} v=0, \quad V_{m}(v)=0 \quad(m=1,2,3,4) \tag{18}
\end{equation*}
$$

where the adjoint differential operator $L^{*}$ is related to $L_{k n}$ by

$$
\begin{equation*}
\int_{y_{1}}^{y_{2}}\left\{v L_{k n}\left(\phi^{[k ; n]}\right)-\phi^{[k ; n]} L^{*} v\right\} d y=\left[p\left(\phi^{[k ; n]}, v\right)\right]_{y_{2}}^{y_{2}} . \tag{19}
\end{equation*}
$$

Here the right-hand member is an ordinary bilinear form and thus reducible to a canonical form

$$
[p]_{y_{1}}^{y_{2}}=U_{1} V_{8}+U_{2} V_{7}+\ldots+U_{8} V_{1}
$$

in which $U_{m}(m=1,2, \ldots, 8)$ are any linearly independent homogeneous expressions in $\phi^{[k ; n]}$ and its derivatives up to the third order evaluated at the end-points $y_{1}$ and $y_{2}$. In particular, $U_{1}, U_{2}, U_{3}$ and $U_{4}$ can be taken to be the left side of the boundary conditions of the non-homogeneous system (17). Once $U_{1}$ to $U_{8}$ are chosen, $V_{1}$ to $V_{8}$ are uniquely determined by the above equation. Hence, if $v$ is the eigensolution of the adjoint system (18) and satisfies

$$
\begin{equation*}
\int_{y_{1}}^{y_{2}} v\left[i \alpha c^{[n-1]} G \delta_{k 1}+H_{k n}\right] d y=\gamma_{1} V_{8}+\gamma_{2} V_{7}+\gamma_{3} V_{6}+\gamma_{4} V_{5}, \tag{20}
\end{equation*}
$$

then the system (17) may have a solution and $c^{[n-1]}$ is given by

$$
\begin{equation*}
i \alpha c^{[n-1]} \int_{y_{1}}^{y_{2}} v G d y=\gamma_{1} V_{8}+\ldots+\gamma_{4} V_{5}-\int_{y_{1}}^{y_{2}} v H_{1 n} d y . \tag{21}
\end{equation*}
$$

It should be pointed out that the adjoint system is assumed to possess eigensolutions. However, this is true only when the associated homogeneous system possesses eigensolutions.

## 3. Solution of the problem

A brief outline of the solution to the above-formulated problem will now be given.

First consider the component with $k=1$ and $n=1$. Substituting these values of $k$ and $n$ into ( 8 ), one obtains the equation governing the amplitude variation function $\phi^{(1 ; 1)}$ associated with the basic harmonic of order $A$

$$
\left\{\left(a^{(0)}+i b^{(0)}+i \alpha \bar{u}\right)\left(D^{2}-\alpha^{2}\right)-i \alpha D^{2} \bar{u}-R^{-1}\left(D^{2}-\alpha^{2}\right)^{2}\right\} \phi^{(1 ; 1)}=0 .
$$

The boundary condition at the fixed wall is simply

$$
\phi^{(1 ; 1)}=D \phi^{(1 ; 1)}=0 \quad \text { at } \quad y=1
$$

and from (A 2) and (A3) with (15) and (A 1) the free surface boundary conditions are found to be

$$
\begin{gathered}
D^{2} \phi^{(1 ; 1)}+\left[\alpha^{2}-3 \alpha /\left(-b^{(0)}+i a^{(0)}-\alpha \bar{u}(0)\right)\right] \phi^{(1 ; 1)}=0, \\
{\left[\alpha^{2}\left(3 \cot \beta+\alpha^{2} S R\right) /\left(-b^{(0)}+i \alpha^{(0)}-\alpha \bar{u}(0)\right)\right] \phi^{(1 ; 1)}+\left[R\left(-b^{(0)}+i a^{(0)}-\alpha \bar{u}(0)\right)\right.} \\
\left.\quad+3 i \alpha^{2}\right] D \phi^{(1 ; 1)}-i D^{3} \phi^{(1,1)}=0 \quad \text { at } \quad y=0 .
\end{gathered}
$$

If one puts

$$
b^{(0)}=-\alpha c_{r} \quad \text { and } \quad a^{(0)}=\alpha c_{i}
$$

in the above differential system, one recovers the Orr-Sommerfeld problem for the film flow which was formulated by Yih (1954). The solution of this problem was first correctly obtained by Benjamin (1957) up to the first order in small $\alpha R$. The solution accurate to the order ( $\alpha R)^{2}$ can be easily obtained with Yih's (1963) method. The eigenfunction $\phi^{(1 ; 1)}$ obtained from a homogeneous system is determined only up to an arbitrary constant multiplier.

For $n=1, k=0$, one has the system

$$
\begin{gathered}
\left(\alpha R c_{i}-D^{4}\right) \phi^{(0 ; 1)}=0, \\
\phi^{(0 ; 1)}=D \phi^{(0 ; 1)}=0 \quad \text { at } \quad y=1, \\
D^{2} \phi^{(0 ; 1)}=\phi^{(0 ; 1)}=0 \quad \text { at } \quad y=0 .
\end{gathered}
$$

With $c_{i}$ given as the imaginary part of the eigenvalue in the linearized theory, the above system has only a trivial solution. All other components of order $A$ (i.e. $k>1$ ) vanish according to our expansion formalism.

Similarly, for $n=2$, only waves with harmonics lower than the second (i.e. $k \leqslant 2$ ) can be of order $A^{2}$. Thus, for $n=2$, the only values of $k$ that need to be considered are 2, 1 and 0 . From (8) with $n=2, k=1$, one has

$$
\begin{gather*}
i\left[\left(-2 i a^{(0)}+b^{(0)}+\alpha \bar{u}\right)\left(D^{2}-\alpha^{2}\right)+3 \alpha\right] \phi^{(1 ; 2)}-R^{-1}\left(D^{2}-\alpha^{2}\right)^{2} \phi^{(1 ; 2)} \\
=i \alpha c^{[1]}\left(D^{2}-\alpha^{2}\right) \phi^{(1 ; 1)} . \tag{22}
\end{gather*}
$$

The corresponding boundary conditions can be obtained again from (12), (A2) and (A 3). It can be shown that $c^{[1]}=0$. The solution $\phi^{(1 ; ~ 2)}$ is then readily obtained.

Similarly, $\phi^{(0 ; 2)}$ and $\phi^{(2 ; 2)}$ can be obtained. The computation is lengthy but straightforward and thus its detail will be omitted.

Finally, to obtain the second Landau coefficient, one considers the component with $k=1$ and $n=3$. From (8), one has the following equation which governs $\phi^{(1 ; 3)}$ :

$$
\begin{equation*}
L_{13} \phi^{(1 ; 3)}=i \alpha c^{[2]} G^{\prime}+H_{13}^{\prime}, \tag{23}
\end{equation*}
$$

where

$$
\begin{gathered}
L_{13}=-\left(D^{2}-\alpha^{2}\right)^{2}+i R\left[\left(-3 i a^{(0)}+b^{(0)}+\alpha \bar{u}\right)\left(D^{2}-\alpha^{2}\right)-\alpha D^{2} \bar{u}\right] \\
G^{\prime}=R\left(D^{2}-\alpha^{2}\right) \phi^{(1 ; 1)}, \\
H_{13}^{\prime}=i \alpha R\left[-D \phi^{(0 ; 2)}\left(D^{2}-\alpha^{2}\right) \phi^{(1 ; 1)}-D \bar{\phi}^{(; ; 2)}\left(D^{2}-\alpha^{2}\right) \phi^{(1 ; 1)}\right. \\
\left.+D \phi^{(2 ; 2)}\left(D^{2}-\alpha^{2}\right) \bar{\phi}^{(1 ; 1)}-\bar{\phi}^{(1 ; 1)} D\left(D^{2}-4 \alpha^{2}\right) \phi^{(2 ; 2)}\right] .
\end{gathered}
$$

From (13), one has the boundary condition at the fixed floor

$$
\begin{equation*}
\phi^{(1 ; 3)}=D \phi^{(1 ; 3)}=0 \quad \text { at } \quad y=1 \tag{24}
\end{equation*}
$$

From (A2) and (A3) one has the vanishing of the shear stress at the free surface

$$
\begin{equation*}
D^{2} \phi^{(1 ; 3)}+\left[\alpha^{2}-3 /\left(c^{\prime}+2 i c_{i}\right)\right] \phi^{(1 ; 3)}=\gamma_{3} \quad \text { at } \quad y=0 \tag{25}
\end{equation*}
$$

and the condition on normal forces at the free surface

$$
\begin{equation*}
D^{3} \phi^{(1 ; 3)}+i \alpha R\left(c^{\prime}+2 i c_{i}+3 i \alpha / R\right) D \phi^{(1 ; 3)}+\frac{i \alpha R Q}{c^{\prime}+2 i c_{i}} \phi^{(1 ; 3)}=\gamma_{4} \quad \text { at } \quad y=0 \tag{26}
\end{equation*}
$$

where $\gamma_{3}$ and $\gamma_{4}$ are known functions of order $1 / \alpha R$.
To obtain $c^{[2]}$ from (21) with $\gamma$ s given on the right sides of the boundary conditions, one still needs the solution $v$ of a homogeneous system which is adjoint to (24) with (25) and (26). To obtain $v$ one rewrites $L_{13}$ in the following form:

$$
L_{13}=-D^{4}+p_{2} D^{2}+p_{4},
$$

where

$$
\begin{gathered}
p_{2}=2 \alpha^{2}-i \alpha R\left[c+2 i c_{i}-\frac{3}{2}\left(1-y^{2}\right)\right], \\
p_{4}=i \alpha^{3} R\left[c+2 i c_{i}-\frac{3}{2}\left(1-y^{2}\right)+3 i \alpha R-\alpha^{4}\right] .
\end{gathered}
$$

The adjoint differential operator $L_{13}^{*}$ is easily found to be (Ince 1926)

$$
L_{13}^{*}=-D^{4}+D^{2} p_{2}+p_{4} .
$$

$L_{13}$ and $L_{13}^{*}$ are related by the relation given by (19) with $k=1, n=3$. For the present problem, the right side of (19) is simply

$$
P=\phi\left[-D\left(p_{2} v\right)+D^{3} v\right]+D \phi\left[p_{2} v-D^{2} v\right]+D^{2} \phi[D v]-D^{3} \phi[v],
$$

in which $\phi$ stands for $\phi^{(1 ; 3)}$.
Thus one has

$$
\begin{align*}
{[p]_{0}^{1}=\phi_{1} v_{1}^{\prime \prime \prime}-} & p_{2}(1) \phi_{1} v_{1}^{\prime}+3 i \alpha R \phi_{1} v_{1}+p_{2}(1) \phi_{1}^{\prime} v_{1}-v_{1}^{\prime \prime} \phi_{1}^{\prime}+\phi_{1}^{\prime \prime} v_{1}^{\prime}-v_{1} \phi_{1}^{\prime \prime \prime} \\
& -\phi_{0} v_{0}^{\prime \prime \prime}+p_{2}(0) \phi_{0} v_{0}^{\prime}-p_{2}(0) \phi_{0}^{\prime} v_{0}+v_{0}^{\prime \prime} \phi_{0}+v_{0}^{\prime \prime} \phi_{0}^{\prime}+v_{0} \phi_{0}^{\prime \prime \prime} \tag{27}
\end{align*}
$$

where primes denote differentiation and the subscript 0 or 1 denotes the endpoint $y=0$ or $y=1$ where a function is to be evaluated. It can be shown that
the right side of the above equation is an ordinary bilinear form and thus is reducible to a canonical form

$$
U_{1} V_{8}+U_{2} V_{7}+\ldots+U_{8} V_{1},
$$

where the $U$ s are any linearly independent homogeneous algebraic expressions in $\phi^{[1 ; 3]}$ and their derivatives up to the third order evaluated at end-points. Here one takes $U_{1}, U_{2}, U_{3}$ and $U_{4}$ to be the left sides of the four boundary conditions given by (24), (25) and (26), and $U_{5}, U_{6}, U_{7}$ and $U_{8}$ will be taken in such a way that all $U \mathrm{~s}$ are linearly independent. For example, $U_{5}=\phi_{0}, U_{6}=\phi_{0}^{\prime}, U_{7}=\phi_{1}^{\prime \prime}$ and $U_{8}=\phi_{1}^{\prime \prime \prime}$. Once $U_{\mathrm{s}}$ are so chosen $V_{n}(n=1,2, \ldots, 8)$ are uniquely determined by (27). They are found to be

$$
\left.\begin{array}{l}
V_{1}=-v_{1}, \quad V_{2}=v_{1}^{\prime}, \quad V_{3}=v_{0}^{\prime \prime}+\alpha^{2} v_{0},  \tag{28}\\
V_{4}=-v_{0}^{\prime \prime \prime}+\left[\alpha^{2}-i \alpha R\left(c^{\prime}+2 i c_{i}\right)+3 /\left(c^{\prime}+2 i c_{i}\right)\right] v_{0}^{\prime}-\frac{i \alpha R Q}{c^{\prime}+2 i c_{i}} v_{0}, \\
V_{5}=v_{0}, \quad V_{6}=-v_{0}^{\prime}, \\
V_{7}=\left[2 \alpha^{2}-i \alpha R\left(c^{\prime}+2 i c_{i}\right)\right] v_{1}-v_{1}^{\prime \prime}, \\
V_{8}=v_{1}^{\prime \prime \prime}-\left[2 \alpha^{2}-i \alpha\left(c^{\prime}+2 i c_{i}\right)\right] v_{1}^{\prime}+3 i \alpha R v_{1} .
\end{array}\right\}
$$

Thus, the adjoint system is found to be

$$
\begin{equation*}
L_{13}^{*} v=0, \quad V_{n}=0 \quad(n=1,2,3,4), \tag{29}
\end{equation*}
$$

with $V_{n}$ given by the first four equations in (28). If this adjoint system possesses a non-trivial solution then ${ }^{[2]}$ has to be obtained from (21).

The first-order solution of (29) is non-trivial and is given by

$$
\begin{equation*}
v=(1-3 y / 2)-\left(y^{3} / 2\right) . \tag{30}
\end{equation*}
$$

Fortunately, only this accuracy is needed for the evaluation of $c^{[2]}$. This can be seen from the following order-of-magnitude argument. Observe that $\gamma_{1}=0$, $\gamma_{2}=0$ and $\gamma_{3}$ as well as $\gamma_{4}$ is of order $1 / \alpha R$, while $V_{5}$ and $V_{6}$ are of order 1. Hence the right side of $(20)$ is of order $1 / \alpha R$. On the other hand, $H_{13}^{\prime}$ is of order $\alpha R$ and $G^{\prime}$ is of order 1 , and $c^{[2]}$ will be found afterwards to be of order 1 . Therefore the left side of (20) is of order $\alpha R$. To neglect a term of order $\alpha R$ in $v$ amounts to neglecting a term of order $(\alpha R)^{2}$ compared with a term of order $1 / \alpha R$ in (20). Thus $c^{[2]}$ obtained from (20) or (21) with $v$ given by (30) is significant up to order $(\alpha R)^{2}$.

Substituting the known functions $H_{13}^{\prime}, G^{\prime}, v$ and the known constants $\gamma_{3}, \gamma_{4}$ $\left(\gamma_{1}=\gamma_{2}=0\right), V_{5}$ and $V_{6}$ into (21), one has from its real and imaginary parts (after some algebra),

$$
\left.\begin{array}{l}
-\alpha R(5 / 4+4 \cot \beta / 3 R) c_{i}^{[2]}-2 c_{r}^{[2]}=R_{1}  \tag{31}\\
-2 c_{i}^{[2]}+\alpha R(5 / 4+4 \cot \beta / 3 R) c_{r}^{[2]}=R_{2},
\end{array}\right\}
$$

where

$$
\begin{aligned}
R_{1}= & 4 a_{0 r}-2 a_{1 r}+23 c_{i} a_{0 i} / 12 \\
& +\alpha R\left[(7 Q / 36+8 \cot \beta / 3 R-6) a_{0 i}-(4 \cot \beta / 3 R-5 / 4) a_{1 i}-7 a_{2 i} / 5\right], \\
R_{2}= & 4 a_{0 i}-2 a_{i r}-23 c_{i} a_{0 r} / 12 \\
& \left.-\alpha R[7 Q / 36+8 \cot \beta / 3 R-6) a_{0 r}-(4 \cot \beta / 3 R-5 / 4) a_{1 r}-7 a_{2 r} / 5\right]
\end{aligned}
$$

in which the subscript $r$ or $i$ denotes the real or imaginary part of the integration constants for $\phi^{(2,2)}$. Solution of (31) gives

$$
\begin{align*}
c_{i}^{[2]}=- & \frac{\alpha R}{D}\left[\left\{\frac{8 S}{R^{2}}-\frac{4 c_{i}}{N^{3}}\right\}\left\{8-\frac{23}{24} \frac{c_{i}}{N}\left(\frac{17}{5}+\frac{22 Q}{9}\right)\right\}\right. \\
& +\left(\frac{5}{16}+\frac{\cot \beta}{3 R}\right)\left(16 S \alpha^{2}-\frac{8 c_{i}}{N}\right)\left(\frac{16}{3 R} \cot \beta-\frac{7}{4} \frac{c_{i}}{N}+\frac{59 Q}{6}\right)+\left(\frac{8 c_{i}}{N}-\frac{152 Q}{9}\right) W \\
& +2 \alpha^{2} S\left\{\left(\frac{34}{5}+\frac{44 Q}{9}+\frac{8 c_{i}}{3 N}\right)\left(\frac{37}{6} \frac{c_{i}}{N}+\frac{67 Q}{36}-\frac{8 \cot \beta}{R}\right)\right. \\
& \left.\left.+\left(\frac{4 \cot \beta}{3 R}-\frac{5}{4}\right)\left(\frac{11}{3} \frac{c_{i}}{N}-\frac{121 Q}{9}\right)-\frac{56}{15} \frac{c_{i}}{N}\right\}\right] \tag{32}
\end{align*}
$$

and

$$
\begin{align*}
c_{r}^{[2]}=\frac{1}{D} & {\left[N ^ { 2 } ( \frac { 5 } { 1 6 } + \frac { \operatorname { c o t } \beta } { 3 R } ) \left\{8\left(\frac{2 S}{R^{2}}-\frac{c_{i}}{N^{3}}\right)\left[8-\frac{23}{24} \frac{c_{i}}{N}\left(\frac{17}{5}+\frac{22 Q}{9}\right)\right]\right.\right.} \\
& -W\left(\frac{118 Q}{3}+\frac{64 \cot \beta}{3 R}+\frac{2 c_{i}}{3 N}\right) \\
& +4 \alpha^{2} S\left[\left(\frac{34}{5}+\frac{44 Q}{9}+\frac{8 c_{i}}{3 N}\right)\left(\frac{37 c_{i}}{6 N}+\frac{67 Q}{36}-\frac{3 \cot \beta}{R}\right)\right. \\
& \left.\left.+\left(\frac{4 \cot \beta}{3 R}-\frac{5}{4}\right)\left(\frac{1 c_{i}}{3 N}-\frac{101 Q}{9}\right)-\frac{56 c_{i}}{15 N}\right]\right\}-16 W \\
& -\left(8 \alpha^{2} S-\frac{4 c_{i}}{N}\right)\left(\frac{16}{3 R} \cot \beta-\frac{7 c_{i}}{4 N}+\frac{59 Q}{6}\right) \\
& -N^{2} W\left\{-\left(\frac{17}{5}+\frac{22 Q}{9}+\frac{4 c_{i}}{N}\right)\left(\frac{7 Q}{36}+\frac{8 \cot \beta}{3 R}-6\right)\right. \\
& \left.\left.+\left(\frac{27}{5}-\frac{64 Q}{9}-\frac{8 c_{i}}{N}\right)\left(\frac{4 \cot \beta}{3 R}-\frac{5}{4}\right)+\frac{7}{5}\left(\frac{17}{5}+\frac{22 Q}{9}+\frac{8 c_{i}}{3 N}\right)\right\}\right] \tag{33}
\end{align*}
$$

where $N=\alpha R$,

$$
\begin{aligned}
W & =805 Q / 2352-437 c_{i} / 112 N-11 \alpha^{2} S / 7-48 / R^{2} \\
D & =N^{2} W^{2}+\left(4 S \alpha^{2}-2 c_{i} / N\right)^{2}
\end{aligned}
$$

Landau's second coefficient $a^{[2]}$, being defined as $\alpha c_{i}^{[2]}$, thus depends on surface tension, the angle of inclination of the plane, the Reynolds number and wavelength according to (32). It is observed that $a^{[2]}$ is not always negative. For example, for values of $\beta=90^{\circ}, S=0$ and $c_{i}$ of order ( $\left.\alpha R\right)^{3}$, the first term in (32) is dominant and $a^{[2]}$ is thus positive. Hence one concludes that for a liquid with zero surface tension a supercritically stable wave motion in a vertically falling film is impossible. However, such a wave motion is possible for liquids with sufficiently large surface tension, as the following numerical examples will show. It is seen from (5) that, for a flow with a negative value of $a^{[2]}$ in a supercritical region ( $c_{i}>0$ ), the exponential growth of the initially infinitesimal wave is modified according to the following equation:

$$
\frac{d A}{d t}=\alpha c_{i} A+a^{[2]} A^{3}
$$

where higher-order terms in (5) are neglected. The general solution of the above equation is easily obtained as (Schade 1964)

$$
A^{2}=\frac{\alpha c_{i} \exp \left[2 \alpha c_{i}\left(t-t_{0}\right)\right]}{1-a^{[2]} \exp \left[2 \alpha c_{i}\left(t-t_{0}\right)\right]},
$$

in which $t_{0}$ is a constant of integration which only changes the origin of the time axis. Thus, as $t$ goes from $-\infty$ to $+\infty$, the amplitude increases from zero to a constant value

$$
\begin{equation*}
A=-\alpha c_{i} / a^{[2]} \tag{34}
\end{equation*}
$$

On the other hand, the speed of the initial wave is modified according to (6) and attains a constant wave speed

$$
\begin{equation*}
\bar{c}=\left(c_{r}+A^{2} c_{r}^{[2]}\right) \bar{u}_{a}, \tag{35}
\end{equation*}
$$

as the amplitude attains the constant value given by (34).
The above results will now be compared with Kapitza's (1949) experimental results. Kapitza observed a stable wave motion on a vertically falling water film at $15^{\circ} \mathrm{C}$. The following quantities were measured: $d=0.013 \mathrm{~cm}, A=0.16 d$ (from figure 1.3 on page 704 of Kapitza's collected work), wavelength $\lambda=0.89 \mathrm{~cm}$, $\bar{c}=12.4 \mathrm{~cm} / \mathrm{sec}, d \bar{u}_{a}=0.061 \mathrm{~cm}^{3} / \mathrm{sec}$, surface tension $\sigma=74 \mathrm{dyne} / \mathrm{cm}$. From these data one obtains $\alpha=0.092, c_{i}=0.2325, R=5.38, N=0.495, S=258.3$, $Q=2 \cdot 2$. Substituting these values into (32), one finds $c_{i}^{[2]}=-9 \cdot 69$ and hence, from (34), $A=0.156 d$, which compares very well with the measured value of $0 \cdot 16 d$. Similarly, one finds $c_{r}^{[2]}=1 \cdot 11$ from (33), and from (35) $\bar{c}$ is found to be $3 \cdot 027 \bar{u}_{a}$, which is considerably larger than the measured value of $2 \cdot 64 \bar{u}_{a}$. Kapitza also used alcohol in his experiment. The measured quantities are: $d=0.0162 \mathrm{~cm}$, $A=0.17 d, \lambda=0.71 \mathrm{~cm}, d \bar{u}_{a}=0.06 \mathrm{~cm}^{3} / \mathrm{sec}, \sigma=29 \mathrm{dyne} / \mathrm{cm}, \bar{c}=10.7 \mathrm{~cm} / \mathrm{sec}$. From these data, one obtains $\alpha=0.1438, c_{i}=0.26, R=3.35, N=0.481$, $S=96 \cdot 0, Q=1 \cdot 98$. Substituting these values into (32) and (33), one finds $c_{i}^{[2]}=-9 \cdot 39$ and $c_{r}^{[2]}=3 \cdot 26$ and then, from (34) and (35), $A=0 \cdot 163 d, \bar{c}=3 \cdot 086 \bar{u}_{a}$. Again, calculated amplitude $A=0 \cdot 163 d$ compares very well with the measured value of $A=0 \cdot 17 d$ but the calculated wave speed $\bar{c}=3.086 \bar{u}_{a}$ is larger than the experimental results of $\bar{c}=2 \cdot 5 \bar{u}_{a}$. It is very puzzling that the speed of finiteamplitude wave motion observed by Kapitza is always smaller than the speed of the infinitesimal surface wave, which is known to be equal to $3 \bar{u}_{a}$. Kapitza's observation is in fact contradictory to Binnie's (1957) observation. In Binnie's experiment, it was found that the speed of the finite-amplitude wave is greater than $3 \bar{u}_{a}$. The following quantities were measured by Binnie for a water film at $19^{\circ} \mathrm{C}: d=4.4 \times 10^{-3} \mathrm{in}$., $\lambda=0.45 \mathrm{in}$., the flow rate $=6.9 \times 10^{-3} \mathrm{in} .{ }^{3} / \mathrm{sec}, \bar{c}=5.5$ in. $/ \mathrm{sec}$. From these values and using $\sigma=0.00499 \mathrm{Lb} . / \mathrm{ft} ., \nu=1.09 \times 10^{-5} \mathrm{ft} .2 / \mathrm{sec}$, one obtains $\alpha=0.0615, R=4.4, N=0.271, \bar{c}=3.51 \bar{u}_{a}, c_{i}=0.1815$ and $Q=1 \cdot 59$. On the substitution of these values into (32), (33) and (34), one finds $A=0.145 d$ and $c_{r}^{[2]}=0.825$. Unfortunately, the amplitude was not recorded in Binnie's experiment and a direct comparison is not possible. However, if one substitutes this value of $A$ and $c_{r}^{[2]}=0.825$ into (35), one has $\bar{c}=3.0173 \bar{u}_{a}$, which is smaller than the observed value of $3.5 \mathrm{l} \bar{u}_{a}$. Thus the present theoretical result predicts a greater wave speed than that observed by Kapitza but a smaller wave speed than that observed by Binnie.

The comparison between the theory and Kapitza's observation has been made only for two experimental points. The comparison for other experimental points is not possible either because the observed waves are so short that $\alpha R$ is no longer small and the theory does not apply or because an accurate reading of the film thickness from Kapitza's photographs even with the aid of a microscope is not possible.

## 4. Conclusion

A closed-form solution for the non-linear development of an initially infinitesimal periodic two-dimensional disturbance is given. It is shown that supercritically stable wave motion is possible in a viscous film. The dependence of the amplitude and the propagation speed of such stable waves on the surface tension of the fluid, the Reynolds number of the flow, the wavelength and the angle of inclination is given explicitly. The agreement between the theoretically predicted wave amplitude and the observed values is excellent. The unsatisfactory agreement between theory and observation on the wave speed is conjectured to be due to the presence of surface contamination in the experiments. To verify this conjecture a rigorous analysis, together with a careful experiment on the effects of surface-active agents, is necessary.

## Appendix

In the following equations, primes denote differentiation with respect to $y$ and a bar denotes complex conjugate. All functions are evaluated at $y=0$ and $k \geqslant 1$.

$$
\begin{aligned}
& \text { The recurrence relation for } \eta^{(k ; n)} \\
& -\left[n \alpha c_{i}-i k \alpha c_{r}+i k \alpha \bar{u}(0)\right] \eta^{(k ; n)} \\
& =i k \alpha \phi^{(k ; n)}+i \alpha\left[(k-m) \phi^{\prime(k-m ; n-p)} \eta^{(m ; p)}+(k+m) \phi^{\prime(k+m ; n-p)} \bar{\eta}^{(m ; p)}\right. \\
& \left.-m \bar{\phi}^{\prime(m ; p)} \eta^{(k+m ; n-p)}\right]+i \alpha\left[\frac { k - m } { 1 + \delta _ { m o } } \phi ^ { \prime \prime ( k - m ; n - p - q ) } \left(\eta^{(m-j ; p)} \eta^{(; ; q)}\right.\right. \\
& \left.+2 \bar{\eta}^{(i ; p)} \eta^{(m+j ; q)}\right)+\frac{k+m}{1+\delta_{m o}} \phi^{\prime \prime(k+m ; n-p-q)}\left(\bar{\eta}^{(m-j ; p)} \bar{\eta}^{(j ; q)}+2 \bar{\eta}^{(m+j ; p)} \eta^{(i ; q)}\right) \\
& \left.-\frac{m}{1+\delta_{(k+m) o}} \bar{\phi}^{n(m ; p)}\left(\eta^{(k+m-j ; n-p-q)} \eta^{(i ; q)}+2 \bar{\eta}^{(j ; n-p-q)} \eta^{(k+m+j ; q)}\right)\right] / 2 \\
& +\left[m a^{[n-m]}+i k b^{[n-m]}\right] \eta^{\{k ; m\}}+\delta_{k l}\left[a^{[n-1]}+i k b^{[n-1]}\right] \eta^{[k ; l]} \\
& +i \alpha\left[j \phi^{\prime(k-j ; n-m]} \eta^{(j ; m)}+(k+j) \bar{\phi}^{\prime j ; m]} \eta^{(k+j ; n-m)}-j \bar{\eta}^{(j ; m)} \phi^{[k+j ; n-m\}}\right] \\
& +i \alpha\left[\frac{k-j}{1+\delta_{j o}} \eta^{(k-j ; n-p-q)}\left(\phi^{\prime \prime(j-m ; p)} \eta^{(m ; q)}+\phi^{\prime \prime(j+m ; p)} \bar{\eta}^{(m ; q)}+\bar{\phi}^{m(m ; p)} \eta^{(j+m ; q)}\right)\right. \\
& -\frac{j}{1+\delta_{(k+j) o}} \bar{\eta}^{(\dot{j} n-p-q)}\left(\phi^{\prime \prime(k+j-m ; p)} \eta^{(m ; q)}+\phi^{\prime \prime(k+j+m ; p)} \bar{\eta}^{(m ; q)}+\bar{\phi}^{\prime(m ; p)} \eta^{(k+j+m ; q)}\right) \\
& \left.+\frac{k+j}{1+\delta_{j o}} \eta^{(k+j ; n-p-q)}\left(\bar{\phi}^{\prime \prime(j-m ; p)} \bar{\eta}^{(m ; q)}+\phi^{\prime \prime(m ; p)} \bar{\eta}^{(j+m ; q)}+\bar{\phi}^{\prime \prime(j+m ; p)} \eta^{(m ; q)}\right)\right]
\end{aligned}
$$

$$
\begin{align*}
& -3 i \alpha\left[\frac{k}{1+\delta_{m o}} \eta^{(k-m ; n-p-q)}\left(\eta^{(m-j ; p)} \eta^{(j ; q)}+\bar{\eta}^{(j ; p)} \eta^{(m+j ; q)}+\eta^{(m+j ; p)} \bar{\eta}^{(j ; q)}\right)\right. \\
& -\frac{m}{1+\delta_{(k+m) o}} \bar{\eta}^{(m ; n-p-q)}\left(\eta^{(k+m-j ; p)} \eta^{(i ; q)}+\bar{\eta}^{(j ; p)} \eta^{(k+m+j ; q)}+\eta^{(k+m+j ; p)} \bar{\eta}^{(; ; q)}\right) \\
& \left.+\frac{k+m}{1+\delta_{m o}} \eta^{(k+m ; n-p-q)}\left(\bar{\eta}^{(m-j ; p)} \bar{\eta}^{(j ; q)}+2 \bar{\eta}^{(m+j ; p)} \eta^{(j ; q)}\right)\right] / 2+O\left(\eta^{3}\right) . \tag{A1}
\end{align*}
$$

By use of the above formula, the following relations can be easily verified.

$$
\begin{aligned}
\eta^{(1 ; 1)}= & 2 \phi^{(1 ; 1)} / 3, \quad \eta^{(1 ; 2)}=\phi^{(1 ; 2)} / c^{\prime} . \\
\eta^{(2 ; 2)}= & \phi^{(2 ; 2)} / c^{\prime}+\phi^{(1 ; 1)} \phi^{(1 ; 1)} /\left(c^{\prime}\right)^{2} . \\
\eta^{(1 ; 3)}= & {\left[\phi^{(1 ; 3)}+\phi^{\prime 2 ; 2)} \bar{\eta}^{(1 ; 1)}+\bar{\phi}^{\prime(1 ; 1)} \eta^{(2 ; 2)}+\eta^{(; 1)}\left(\phi^{\prime \prime(1 ; 1)} \bar{\eta}^{(1 ; 1)}\right.\right.} \\
& \left.+\bar{\phi}^{\prime \prime(1 ; 1)} \eta^{(1 ; 1)}\right)-c^{(2]} \eta^{(1 ; 1)}+\eta^{(1 ; 1)}\left(\phi^{(0 ; 2)}+\bar{\phi}^{(0 ; 2)}\right) \\
& \left.-3 \eta^{(1 ; 1)} \bar{\eta}^{(1 ; 1)} \eta^{(1 ; 1)} / 2\right] /\left(c^{\prime}+2 i c_{i}\right) .
\end{aligned}
$$

The tangential boundary conditions at the free surface

$$
\begin{align*}
& k^{2} \alpha^{2} \phi^{(k ; n)}+\frac{\alpha^{2}}{1+\delta_{k o}}\left[(k-m)^{2} \phi^{\prime(k-m ; n-q)} \eta^{(m ; q)}+(k+m)^{2} \phi^{\prime(k+m ; n-q)} \bar{\eta}^{(m ; q)}\right. \\
& \left.+m^{2} \bar{\phi}^{\prime}(m ; n-q) \eta^{(k+m ; q)}\right]+\frac{\alpha^{2}}{2\left(1+\delta_{k o}\right)}\left[\frac { ( k - m ) ^ { 2 } } { 1 + \delta _ { m o } } \phi ^ { \prime \prime ( k - m ; n - q - r ) } \left(\eta^{(m-j ; q)} \eta^{(; ; r)}\right.\right. \\
& \left.+\bar{\eta}^{(j ; q)} \eta^{(m+j ; r)}+\eta^{(m+j ; q)} \bar{\eta}^{(j ; r)}\right)+\frac{(k+m)^{2}}{1+\delta_{m o}} \phi^{\prime \prime(k+m ; n-q-r)}\left(\bar{\eta}^{(m-j ; q} \bar{\eta}^{(; r r)}\right. \\
& \left.+\bar{\eta}^{(m+j ; q)} \eta^{(j ; r)}+\bar{\eta}^{(m+j ; q)} \eta^{(j ; r)}\right)+\frac{m^{2}}{1+\delta_{(k+m) o}} \bar{\phi}^{\prime \prime(m ; p)}\left(\eta^{(k+m-j ; n-p-r)} \eta^{(i ; r)}\right. \\
& \left.\left.+\bar{\eta}^{(j ; n-p-r)} \eta^{(k+m+i ; r)}+\eta^{(k+m+j ; n-p-r)} \bar{\eta}^{(i ; r)}\right)\right] \\
& +\frac{\alpha^{2}}{1+\delta_{k o}}\left[m(k-m) \phi^{\prime(k-m ; n-q)} \eta^{(m ; q)}-m(k+m) \phi^{\prime(k+m ; n-q)} \bar{\eta}^{(m ; q)}\right. \\
& \left.-m(k+m) \bar{\phi}^{\prime(m ; p)} \eta^{(k+m ; n-p)}\right]+\frac{\alpha^{2}}{1+\delta_{k o}}\left[\frac { k - j } { 1 + \delta _ { j o } } \phi ^ { \prime \prime } ( k - j ; n - q - r ) \left\{j \eta^{(j-m ; q)} \eta^{(m ; r)}\right.\right. \\
& \left.+(j+m) \bar{\eta}^{(m ; q)} \eta^{(j+m ; r)}-m \eta^{(j+m ; \varphi)} \bar{\eta}^{(m ; r)}\right\}-\frac{k+j}{1+\delta_{j o}} \phi^{\prime \prime(k+j ; n-q-r)}\left\{m \bar{\eta}^{(j-m ; q)} \bar{\eta}^{(m ; r)}\right. \\
& \left.-m \bar{\eta}^{(j+m ; q)} \eta^{(m ; r)}+(j+m) \bar{\eta}^{(j+m ; q)} \eta^{(m ; r)}\right\}-\frac{j \bar{\phi}^{\prime(j ; p)}}{1+\delta_{(k+j) o}}\left\{m \eta^{(k+j-m ; n-p-r)} \eta^{(m ; r)}\right. \\
& \left.\left.+(k+j+m) \bar{\eta}^{(m ; n-p-r)} \eta^{(k+j+m ; r)}-m \eta^{(k+j+m ; n-p-r)} \bar{\eta}^{(m ; r)}\right\}\right]+\phi^{\prime \prime}(k ; n) \\
& +\frac{1}{1+\delta_{k o}}\left[\phi^{\prime \prime \prime(k-m ; n-q)} \eta^{(m ; q)}+\phi^{\prime \prime \prime(k+m ; n-q)} \bar{\eta}^{(m ; q)}+\bar{\phi}^{\prime \prime(m ; n-q)} \eta^{(k+m ; q)}\right] \\
& +\frac{1}{2\left(1+\delta_{k o}\right)}\left[\frac{\phi^{\mathrm{iv}(k-m ; n-q-r)}}{1+\delta_{m o}}\left(\eta^{(m-j ; \varphi)} \eta^{(; ; r)}+\bar{\eta}^{(; ; \varphi)} \eta^{(m+j ; r)}+\eta^{(m+j ; \varphi)} \bar{\eta}^{(; ; r)}\right)\right. \\
& +\frac{\phi^{i \mathrm{~V}(k+m ; n-q-r)}}{1+\delta_{m o}}\left(\bar{\eta}^{(m-j ; q)} \bar{\eta}^{(; ; r)}+2 \bar{\eta}^{(m+j ; q)} \eta^{(j ; r)}\right)+\frac{\bar{\phi}^{1 \mathrm{~V}(m ; p)}}{1+\delta_{(k+m) a}}\left(\eta^{(k+m-j ; n-p-r)} \eta^{(; ; r)}\right. \\
& \left.\left.+\bar{\eta}^{(j ; q)} \eta^{(k+m+j ; r)}+\eta^{(k+m+j ; n-p-r)} \bar{\eta}^{(j ; r)}\right)\right]+O\left(\eta^{3}\right)=0 . \tag{A2}
\end{align*}
$$

The normal boundary condition at the free surface ( $k \geqslant 1$ )

$$
\begin{align*}
& -p^{(k ; n)}-\left[p^{\prime(k-m ; n-q)} \eta^{(m ; q)}+p^{\prime(k+m ; n-q)} \bar{\eta}^{(m ; q)}+\bar{p}^{\prime(m ; p)} \eta^{(k+m ; n-p)}\right] \\
& -\frac{p^{\prime \prime(k-j ; n-q-r)}}{1+\delta_{j o}}\left[\eta^{(j-m ; q)} \eta^{(m ; r)}+2 \eta^{(j+m ; q)} \bar{\eta}^{(m ; r)}\right] / 2 \\
& -\frac{p^{\prime \prime(k+i ; n-q-r)}}{1+\delta_{j o}}\left[\bar{\eta}^{(j-m ; q)} \bar{\eta}^{(m ; r)}+2 \bar{\eta}^{(j+m ; q)} \eta^{(m ; r)}\right] / 2 \\
& -\frac{\bar{p}^{\prime \prime(j ; n-q-r)}}{1+\delta_{(k+j) o}}\left[\eta^{(k+j-m ; q)} \eta^{(m ; r)}+2 \eta^{(k+j+m ; q)} \bar{\eta}^{(m ; r)] / 2}\right. \\
& -\frac{2 i k \alpha}{R} \phi^{\prime(k ; n)}+\frac{2}{\left(1+\delta_{k o}\right) R}\left[-i \alpha(k-m) \phi^{\prime \prime(k-m ; n-q)} \eta^{(m ; q)}\right. \\
& -i \alpha(k+m) \phi^{\left.\prime \prime(k+m ; n-q) \bar{\eta}^{(m ; q)}+i \alpha m \bar{\phi}^{\prime \prime(m ; p)} \eta^{(k+m ; n-q)}\right]} \\
& +\left[\frac{-i \alpha(k-j)}{1+\delta_{j o}} \phi^{\prime \prime(k-j ; n-q-r)}\left(\eta^{(j-m ; q)} \eta^{(m ; r)}+2 \eta^{(j+m ; q)} \bar{\eta}^{(m ; r)}\right)\right] / R \\
& +\left[\frac{i \alpha(k+j)}{1+\delta_{j o}} \phi^{\prime \prime(k+j ; n-q-r)\left(\bar{\eta}^{(j-m ; q)} \bar{\eta}^{(m ; r)}+2 \bar{\eta}^{(j+m ; q)} \eta^{(m ; r)}\right)}\right] / R \\
& -S k^{2} \alpha^{2} \eta^{(k ; n)}+O\left(\eta^{s}\right)=0 . \tag{A3}
\end{align*}
$$

In the above equation, $p^{[k ; n]}$ is given by (15).

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