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The motions of liquid films in a gas☆

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ABSTRACT

The motions in a gas of thin films of a viscous incompressible liquid acted upon by capillary forces are considered. The surface tension depends on the impurity concentration of a surface-active material, soluble or insoluble in the liquid, and the liquid is non-volatile. The inertia of the liquid, viscous stresses, the Laplace pressure and the surface-tension gradients, impurity transfer and also the particular properties of super-thin films are taken into account. The motions of the films are described using the model of quasi-steady viscous flow. Systems of equations are obtained in the approximation of an ideal compressible medium and for small Mach numbers. The conditions for the incompressible film surface approximation to hold are obtained. The severe limitations of the gas-dynamic approximation in the case of a soluble impurity due to attenuation of the waves related to diffusion are investigated. A continuum model of the film as a compressible medium with a non-equilibrium pressure is constructed. The asymptotic form of the solutions of unsteady problems of impurity transfer in the limit of weak non-equilibrium is obtained. Integrals of the equations of motion of the films in steady one-dimensional problems are derived. Integral forms of the equations of momentum and its moment for an arbitrary contour of the film are presented, which hold for steady flows in a film and in quasi-statics. The boundary conditions for the solutions of the system of equations of motion of films are given.

The finite compressibility of the surface of a film, due to the presence of an impurity, may lead to the film behaving as a compressible two-dimensional medium. It is well known from shallow-water theory,[1] that the presence of a field of mass forces can lead to finite compressibility of a two-dimensional medium. It is also known that in a film of a viscous incompressible liquid, due to the effect of capillary forces, sound waves are possible as in a two-dimensional ideal compressible liquid.[2]

The papers by Gibbs[3] and Taylor,[4] and also the monograph by Mysels, Shinoda and Frenkel[5] are of particular value in the theory of the motions of liquid films when capillary forces are taken into account.

1. A model of quasi-steady viscous flow in a film

We will assume that the surfaces of the film are symmetrical about the $x_3 = 0$ plane in a Cartesian system of coordinates x_1, x_2, x_3. The scale λ of the change in the flow parameters is large compared with the film thickness h: $\lambda \gg h$ and the characteristic time $\lambda \gg h^2/\nu$ is relatively long (ν is the coefficient of kinematic viscosity), so that the flow is close to steady in each section of the film.[6] In the stationary problem, the time τ can be defined in terms of the characteristic velocity. The velocity profile in the cross-section of the film is the superposition of two known velocity profiles with coefficients that vary slowly along x_1 and x_2 (Ref. 6)

$$v_i' = v_i(x_1, x_2, t) + \frac{3}{2}\left(1 - \left(\frac{2x_3}{h}\right)^2\right)u_i(x_1, x_2, t), \quad i = 1, 2 \tag{1.1}$$

Here v_i are the components of the two-dimensional velocity $\boldsymbol{v}$ of the film surface, and u_i are the components of the average velocity $\mathbf{u}$ of the flow of liquid in the film relative to the surface. The equations of the model were written in Ref. 6 taking into account the principal approximation in an inertial term, defined by the velocity $\boldsymbol{v}$. Later, to describe the effect of the second velocity $\mathbf{u}$ on the low attenuation of wave motions of the film, in the inertial term of the equation of momentum the small contribution of $\mathbf{u}$ was taken into account: the velocity $\boldsymbol{v}$ is replaced by the average of $\boldsymbol{v}'$[1] along x_3. We will write

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[1] O.V. Voinov. The principles of two-velocity capillary hydrodynamics of films. Moscow, 1990. Deposited at the VINITI, 4 December 1990, No. 6090-v90.

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the equations of quasi-steady flows in films[6] in the form

$$h\rho\frac{d\overline{\upsilon_i'}}{dt} = 2\nabla_i\sigma + h\rho g_i + h\nabla_i\left(\frac{\sigma}{2}\nabla^2 h - p_e\right) + \nabla_j\{h\mu(2\delta_{ij}\mathrm{div}\boldsymbol{\upsilon} + \nabla_i\upsilon_j + \nabla_j\upsilon_i)\} \tag{1.2}$$

$$6\mu\mathbf{u} = -h\nabla\sigma \tag{1.3}$$

$$\frac{\partial h}{\partial t} + \mathrm{div}(h\bar{\boldsymbol{\upsilon}}') = 0, \quad \bar{\boldsymbol{\upsilon}}' = \boldsymbol{\upsilon} + \mathbf{u} \tag{1.4}$$

$$\sigma = \sigma(\Gamma, T), \quad \Gamma = \Gamma(c, T), \quad x_3 = \pm\frac{1}{2}h \tag{1.5}$$

$$\frac{\partial \Gamma}{\partial t} + \mathrm{div}(\Gamma\boldsymbol{\upsilon}) = -D\frac{\partial c}{\partial x_3}\left(x_1, x_2, \frac{h}{2}, t\right) \tag{1.6}$$

$$\frac{\partial c}{\partial t} + \upsilon_j'\nabla_j c - (x_3\mathrm{div}\boldsymbol{\upsilon})\nabla_3 c = D\nabla^2 c, \quad x_3 \in \left(-\frac{h}{2}, \frac{h}{2}\right) \tag{1.7}$$

The bar denotes an average over the cross-section (the bar over v_i' will henceforth be omitted if it is clear that v_i' does not correspond to the profile (1.1)), summation is carried over the repeated index $j = 1, 2$, g_i are the components of the mass force $\mathbf{g}$, ρ is the density, μ is the dynamic viscosity coefficient, D is the diffusion coefficient, σ is the surface tension with isotherm (1.5), Γ and c are the surface and volume concentrations of impurity, related on the surface by the adsorption isotherm (1.5), and p_e is the additional effective pressure in super-thin films (the disjoining pressure with a minus sign); the temperature T and the gas pressure are constant. Eqs. (1.6) and (1.7) describe the transfer of impurity. The contribution of the rheological properties of the surface layer in the equation of motion is ignored.

The effect of long-range Van-der-Waals forces on the flow of liquid films in a gas and on the solid surface can be described by a unified equation.[2,7–11] There are different ways of deriving it. In Ref. 10 one of the models of the theory of the surface tension of liquids[12] is used. Instead of this, we will consider the equilibrium of a film in the field of an arbitrary mass force $\mathbf{g} = -\nabla U$. The chemical potential of the film includes a term which depends on the thickness,[13,14] and correspondingly a term $p_e(h)$ occurs in the equation of equilibrium of a thin film

$$p_e(h) + \rho U = \mathrm{const} \tag{1.8}$$

where $p_e = -A'/(6\pi h^3)$ and A' is the Hamaker constant (we ignore the delay of the interaction).

We introduce the idea of the longitudinal mass force $\mathbf{g}_e = -\mathbf{g}$, equivalent to the mass force due to molecular Van-der-Waals forces. We then obtain that the equivalent body force $\rho g_e = -\nabla p_g$, which is constant over the cross-section of the film, represents the contribution of Van-der-Waals forces to the hydrodynamics equation.

Hence we see that the continuum description of a film of a Van-der-Waals liquid can be regarded as a consequence of the equation of the statics of a film in a field of a mass force (1.8). This important equation was obtained for the first time by Frenkel[13] (in a somewhat different notation).

2. The boundary conditions for solutions of the system of equations of motion of films

Under actual conditions, free films are often bounded by a relatively thick edge – a meniscus. The dynamics of a film close to the edge were first investigated by Gibbs.[3] We will consider the boundary conditions on the contour of a film for the case of flows with velocities considerably lower than the velocity of sound.[2] We will write the stress tensor in the cross-section of film, subtracting the contribution of the stresses in the gas

$$p_{ij} = \delta_{ij}\left(\frac{1}{2}\sigma\nabla^2 h - p_e\right) + \mu(2\delta_{ij}\mathrm{div}\boldsymbol{\upsilon} + \nabla_i\upsilon_j + \nabla_j\upsilon_i) \tag{2.1}$$

where δ_{ij} is the Kronecker delta. We will divide the arbitrary contour L, which bounds a certain region of the film, as in Gibbs' papers and elsewhere[3,5] (see also O.V. Voinov. Motions of liquid films in a gas under capillary forces. Moscow, 2006. Deposited at the VINITI 17 August 2006, No. 1081-V2006), into a section L_- where the liquid flows into the contour and a section L_+ where the liquid flows out of the contour:

$$\boldsymbol{\upsilon}'\cdot\mathbf{n} < 0, \quad \mathbf{x} \in L_-; \quad \boldsymbol{\upsilon}'\cdot\mathbf{n} > 0, \quad \mathbf{x} \in L_+ \tag{2.2}$$

where $\mathbf{n}$ is the unit vector of the outward normal to L.

We will consider the possible conditions which must be imposed on the components of the velocity or the surface force in the cross-section of the film on the contour L. We will specify the right-hand sides in one of the following three pairs of boundary conditions, usual in the dynamics of a viscous liquid:

$$\boldsymbol{\upsilon}\cdot\mathbf{n} = \upsilon_n(\mathbf{x}, t), \quad \boldsymbol{\upsilon}\cdot\mathbf{l} = \upsilon_l(\mathbf{x}, t), \quad \mathbf{x} \in L \tag{2.3}$$

$$\upsilon_n = \upsilon_n(\mathbf{x}, t), \quad p_l = p_l(\mathbf{x}, t), \quad \mathbf{x} \in L \tag{2.4}$$

$$\mathbf{n}\cdot\mathbf{p}\cdot\mathbf{n} = p_n(\mathbf{x}, t), \quad \mathbf{n}\cdot\mathbf{p}\cdot\mathbf{l} = p_l(\mathbf{x}, t), \quad \mathbf{x} \in L \tag{2.5}$$

Here $\mathbf{1}$ is the unit vector tangential to L.

In addition to one pair of conditions from (2.3)–(2.5) we will specify a further condition, imposed on the film thickness on the inflow line

$$h = h_-(\mathbf{x}, t), \quad \mathbf{x} \in L_- \tag{2.6}$$

The film thickness is not specified on the line where the liquid flows out of the film.

In the case of an insoluble impurity, which can only exist on the surface, we additionally define a line $\tilde{L}_-$ of surface inflow with respect to the normal velocity of the surface $\boldsymbol{v}\cdot\mathbf{n} < 0, \quad \mathbf{x} \in \tilde{L}_-$, and we specify on this line the condition imposed on the surface concentration of impurity

$$\Gamma = \Gamma_-(\mathbf{x}, t), \quad \mathbf{x} \in \tilde{L}_- \tag{2.7}$$

If the impurity is soluble, then instead of condition (2.7) we specify a symmetric profile of the volume concentration of impurity on the contour L

$$c = c(\mathbf{x}, x_3, t), \quad x_3 \in \left(-\frac{h}{2}, \frac{h}{2}\right), \quad \mathbf{x} \in L \tag{2.8}$$

Condition (2.8), imposed on the concentration c, is specified both on the line L_- and on the line L_+.

The shape of the surface with tension, when the normal stress in the liquid is specified on it, is defined by a two-dimensional Poisson equation in h. Conditions either on h or on the normal derivative $\mathbf{n}\cdot\nabla h$ are usually imposed for it, and hence we set one more condition on the outflow line L_+

$$\mathbf{n}\cdot\boldsymbol{\nabla}h = (\nabla_n h)_+(\mathbf{x}, t), \quad \mathbf{x} \in L_+ \tag{2.9}$$

Condition (2.9) is only set when the contribution of the capillary jump of the normal stress $\sigma\nabla^2 h/2$ on the surface of the film is taken into account in the stress tensor p_{ij}, defined in (2.1).

In the case of a non-viscous two-dimensional fluid, when the viscous stresses are not taken into account in the stress tensor (2.1), instead of conditions (2.3)–(2.5), two conditions are specified on the inflow line L_-: the components of the velocity v_n and v_l or the normal stress p_n (the difference between the pressure in the gas and the pressure inside the film) and rot$\boldsymbol{v}$, while on the outflow line L_+ only one condition is specified, namely, the normal velocity v_n or the normal stress p_n.

The mean velocity $\mathbf{u}$ of the liquid relative to the surface does not occur in the boundary conditions written above, since the velocity $\mathbf{u}$ is defined in terms of the field of the quantities σ and h by Eq. (1.3). But the velocity $\mathbf{u}$ must be taken into account when determining the type of line L_- or L_+ according to conditions (2.2), in which $\boldsymbol{v}' = \boldsymbol{v} + \mathbf{u}$.

The function specified on the right-hand sides of boundary conditions (2.3)–(2.9), must satisfy the limits for which the model of the flow in the film holds. In initial boundary-value problems it is also necessary to specify the initial conditions.

3. Integrals and integral forms of the equations of motion of the film

We will introduce the capillary tension tensor s, corresponding to the point on the film surface $x_3 = z(x)$,

$$s_{ij} = \sigma\left(1 + \frac{1}{2}|\nabla z|^2\right)\delta_{ij} - \sigma(\nabla_i z)\nabla_j z, \quad z = \frac{1}{2}h \tag{3.1}$$

Using the identity

$$z\nabla_i\nabla^2 z = \nabla_j\left\{\left(z\nabla^2 z + \frac{1}{2}|\nabla z|^2\right)\delta_{ij} - (\nabla_i z)\nabla_j z\right\}$$

and omitting small quantities of the order of $|\nabla\sigma|h^2\lambda^{-2}$ in the calculations we reduce the momentum Eq. (1.2) to the form[2]

$$h\rho\frac{d\boldsymbol{v}'}{dt} = h\rho\mathbf{g} + \nabla E + \operatorname{div}(h\mathbf{p} + 2\mathbf{s}) \tag{3.2}$$

Here E is the primitive of the effective pressure $p_e(h, T) = \partial E/\partial h$); the quantity $E \to 0$ when $h \to \infty$. Taking the equation of continuity (1.4) into account, we can rewrite Eq. (3.2) in the form

$$\frac{\partial}{\partial t}(h\rho v_i') + \nabla_j I_{ij} = h\rho g_i, \quad I_{ij} = \rho h v_i' v_j' - 2s_{ij} - hp_{ij} - E\delta_{ij} \tag{3.3}$$

For steady flows, when there are no mass forces ($g = 0$), using Eqs (3.3) we can write the following identities for an arbitrary closed contour L, which hold for arbitrary solutions of the equations of motion

$$\int_L I_{ij}n_j dl = 0, \quad \int_L \varepsilon_{3ki}x_k I_{ij}n_j dl = 0, \quad \int_L n v_j' n_j dl = 0 \tag{3.4}$$

ε_{mki} are the components of the Levi-Civita tensor. The first two identities (3.4) also hold for the solutions of non-stationary problems in the non-inertial approximation (in the quasi-statics of films). For plane motion of the film along the x_1 axis, according to the identities (3.4), the quantities I_{11} and hv_1' are constant. In the axisymmetric problem we obtain three integrals of the equations of motion[2]

$$I_{11}x_1 = \text{const}, \quad h v_1' x_1 = \text{const}, \quad x_1^2 I_{\theta 1} = \text{const}$$

Here x_1 is the radius and θ is the angle in polar coordinates.

For the plane case we can write

$$h\rho v'^2 - 4\mu h\nabla_1 v - E + hp_e = 2s_{11} + \frac{1}{2}\sigma\nabla_1^2 h + \text{const} \tag{3.5}$$

where the flow rate $hv' = \text{const}$, and s_{11} is defined by expression (3.1).

The last two terms on the left-hand side of the integral (3.5) are important in the case of superthin films, and the contribution of the capillary forces is on the right-hand side. If we put all the terms on the left-hand side of integral (3.5) equal to zero, we arrive at the Mysels, Shinoda, Frenkel equation,[5] obtained in the non-inertial approximation of the one-dimensional equations of lubrication theory. Integral (3.5) was considered previously in special cases of the non-inertial approximation, taking viscous stresses into account[6] and taking Van-der-Waals forces into account.[8]

4. Integral relation and asymptotic integral in problems of impurity transfer in a film

Consider the transfer of an impurity when the changes in the velocity v along the film are considerably greater than the velocity u (the necessary conditions for this are given below). We put $u = 0$; then $\boldsymbol{v}' = \boldsymbol{v}$. We will also use the condition $\lambda^2 \gg D\tau$ and we will omit the second derivatives with respect to x_1 and x_2 in the transport Eq. (1.7).

We will introduce the mean concentration

$$\bar{c} = \frac{1}{h}\int_{-h/2}^{h/2} c\,dx_3 \tag{4.1}$$

Integrating both sides of the transfer Eq. (1.7) over the cross-section of the film with the above assumptions and using Eqs. (1.4) and (1.6), we obtain

$$h\frac{d\bar{c}}{dt} + 2\frac{d\Gamma}{dt} + 2\Gamma\operatorname{div}\boldsymbol{v} = 0$$

Hence, after integration, taking Eq. (1.4) into account, it follows that

$$\bar{c} + 2\Gamma/h = f(\xi_i) \tag{4.2}$$

Here ξ_i are the Lagrangian coordinates and f is an arbitrary function of ξ_i.[15] The integral relation (4.2) expresses the fact that the mass of impurity in a small element of the film is constant. In the special case when the concentration is constant over the cross-section, relation (4.2) gives the integral obtained earlier in Ref. 8.

Using relation (4.2), we will derive the integral of the non-stationary impurity-transfer problem. Suppose $h^2 \ll D\tau$. Then the concentration c will change only slightly over the cross-section of the film, and the sum of the first two terms of the asymptotic expansion of the concentration c is a quadratic polynomial of x_3. The unknown coefficient of the asymptotic can be found using the integral relation. This mathematical method corresponds to the integral-relation method in the theory of mass transfer in thin films.[3] Correspondingly, we will write the concentration in the asymptotic form

$$c = c_0(x_1, x_2, t) + \frac{3}{2}\left(1 - \left(\frac{2x_3}{h}\right)^2\right)c_1(x_1, x_2, t) \tag{4.3}$$

The following relations are obtained from Eqs. (4.1), (4.3) and (1.6)

$$\bar{c} = c_0 + c_1, \quad c_1 = \frac{h}{6D}\left(\frac{d\Gamma}{dt} + \Gamma\operatorname{div}\boldsymbol{v}\right)$$

Hence also from relation (4.2) we obtain the integral

$$\frac{2\Gamma}{h} + c_0 + \frac{h}{6D}\left(\frac{d\Gamma}{dt} + \Gamma\operatorname{div}\boldsymbol{v}\right) = f(\xi_i) \tag{4.4}$$

where c_0 is the concentration close to the surface and $f(\xi_i)$ is an arbitrary function of the Lagrangian coordinates. Using Eq. (1.4), we

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[2] See O.V. Voinov. Motions of liquid films in a gas under the action of capillary forces. Moscow, 2006. Deposited at the VINITI 17 August 2006, No.1081-v2006.

[3] This method was apparently first mentioned by G.G. Chernyi.

eliminate the velocity from relation (4.4) and obtain the integral

$$\frac{2\Gamma}{h} + c_0 + \frac{h^2}{6D}\frac{d}{dt}\frac{\Gamma}{h} = f(\xi_i) \tag{4.5}$$

The concentrations Γ and c_0 are related by the adsorption equation $\Gamma = \Gamma(c_0, T)$. If we neglect the derivative with respect to time in Eq. (4.5) we obtain the integral obtained previously in Ref. 8.

We will also write Eq. (4.5) in a different form, using the derivative of c_0 with respect to time,

$$\frac{2\Gamma}{h} + c_0 - \frac{h^2}{12D}\frac{dc_0}{dt} = f(\xi_i) \tag{4.6}$$

In relation (4.6), $c_0 = 0$ corresponds to the integral for the case of an insoluble impurity.

Hence, the solution of the problems for transfer Eqs. (1.6) and (1.7) gives one integral (4.5) (or (4.6)), if the condition of weak non-equilibrium $h^2 \ll D\tau$ holds (also for relatively high values of λ and a small value of u).

Using relation (4.5) and taking Eq. (1.5) into account, we can obtain equations describing the surface tension σ.

5. The approximation of an ideal compressible medium when describing film dynamics

We will assume that the effect of impurity on the surface tension is relatively large, i.e. the Gibbs surface elasticity is high and rapid motions of the film are possible in which the inertial term in the momentum Eq. (1.2) is of the order of the surface tension gradient. In such motions, characteristic values of the change in the first velocity $v^* \sim c_s$ are possible, where c_s is the velocity of sound. The characteristic value of the second velocity will then be small: $u \ll v^*$, as can be seen from simple estimates of the terms in the momentum Eq. (1.2) and the equation for the velocity $\mathbf{u}$ (1.3) when the inequality $h^2 \ll \nu\tau$ is taken into account.

If $h^2 \ll D\tau$, the impurity concentration changes very little across the film and the following equilibrium version of integral (4.5) holds

$$2\Gamma/h + c = f(\xi_i) \tag{5.1}$$

Hence, taking Eq. (1.5) into account we can conclude that the surface tension depends on the thickness h and on the Lagrangian coordinates $\sigma = \sigma(h, \xi_i, T)$.[8]

For a high Reynolds number for flow in the film or a similar condition for small perturbations

$$\lambda \upsilon^*/\nu \gg 1, \quad \lambda^2 \gg \nu\tau \tag{5.2}$$

(τ is the characteristic time) the contribution of the viscous stresses in momentum Eq. (1.2) is small. In the limit of long waves, the contribution of the capillary pressure is also small: $\sigma\nabla^2 h/2$. Neglecting small quantities, we obtain the equations of motion of the film – an ideal compressible medium with a two-dimensional pressure $\tilde{p}$:

$$\tilde{p} = -2\sigma + hp_e - E = \tilde{p}(h, \xi_i, T) \tag{5.3}$$

$$\rho h\frac{d\boldsymbol{\upsilon}}{dt} = -\nabla\tilde{p}, \quad \frac{\partial h}{\partial t} + \nabla \cdot h\boldsymbol{\upsilon} = 0 \tag{5.4}$$

The equation of state (5.3) includes the dependence on the initial state, since integral (5.1) includes an arbitrary function of the Lagrangian coordinates.

From relations (5.3)–(5.4) we obtain the velocity of sound c_s in a compressible medium with density ρh:

$$c_s^2 = -\frac{2}{\rho}\frac{\partial\sigma}{\partial h} + \frac{h}{\rho}\frac{\partial p_e}{\partial h} \tag{5.5}$$

Using integral (5.1), we write the coefficient of Gibbs elasticity of the surface $\varepsilon' = s\partial\sigma/\partial s$ (s is the area of a small element of the surface)[8]

$$\varepsilon' = -\Gamma\frac{\partial\sigma}{\partial\Gamma}\left(1 + \frac{h}{2H}\right)^{-1}, \quad H = \frac{\partial\Gamma}{\partial c} \tag{5.6}$$

In the case of an insoluble impurity, the coefficient of elasticity of the surface ε' corresponds to the value $H^{-1} = 0$. The compressibility of the film as a two-dimensional medium is characterized by the coefficient of elasticity

$$K = 2\varepsilon = 2\varepsilon' + h^2\frac{\partial p_e}{\partial h}\left(\frac{\partial\sigma}{\partial h} = -\frac{\varepsilon'}{h}\right) \tag{5.7}$$

Here the elasticity of a two-dimensional medium – the film – has the same meaning as the elasticity of an ideal fluid – a nonlinearly elastic body in continuum mechanics.[16]

From relations (5.5)–(5.7) we have the following formula for the frequency of sound waves with wave number k

$$\omega_0 = c_s k = (2\varepsilon/(\rho h))^{1/2}k \tag{5.8}$$

which agrees, in the case of an insoluble impurity, with the similar formula of linear theory.[2]

We will express the conditions for the model of a two-dimensional compressible medium to hold in terms of the coefficient of elasticity of the surface ε'. The condition for the contribution of the Laplace pressure to be small in the equation of motion takes the form

$$h^2/\lambda^2 \ll \varepsilon'/\sigma \tag{5.9}$$

The following inequality corresponds to condition (5.2) for the longitudinal viscous stresses to be small

$$\mu h/(\tau\varepsilon') \ll 1 \tag{5.10}$$

The effective pressure p_e makes a small contribution to the coefficient of elasticity of the film (5.7) and the velocity of sound (5.8), and we can put $\varepsilon = \varepsilon'$,

$$hp_e/\varepsilon' \ll 1 \tag{5.11}$$

6. Sound waves in a film and the limitations of the applicability of the gas-dynamic approximation

The approximation of an ideal compressible medium may hold true when there is a low energy dissipation in the actual flow. The dissipation is small if small perturbations attenuate slightly. In this connection we will consider small perturbations of the uniform state of the film, periodic along the x axis, for which the thickness has the form

$$h = h_0 + h_1\exp\{i(\omega t - kx)\}, \quad h_1 \ll h_0$$

Note that waves in a thin film differ considerably from linear waves on the surface of a liquid of infinite depth[17] due to the possibility of sound waves in the film.[2]

Suppose the complex frequency ω of the sound wave differs only slightly from the real value ω_0:

$$\omega = \omega_0 + id \tag{6.1}$$

In the case of a soluble impurity, in addition to satisfying the conditions $h^2 \ll \nu\tau$ it is necessary that $h^2 \ll D\tau$, where $\tau \sim 1/\omega_0$. From the equations of motion (1.2)–(1.7) with condition (5.9) we obtain

$$d = 2\nu k^2 + \frac{\varepsilon'^2}{\varepsilon^2}\frac{h^2\omega_0^2}{24\nu} + \frac{\Pr}{W}\frac{h^2\omega_0^2}{24\nu}; \quad \Pr = \frac{\nu}{D}, \quad W = 1 + \frac{2H}{h} \tag{6.2}$$

($W^{-1} = 0$ for an insoluble impurity). The logarithmic decrement d, according to expression (6.2), is due to three energy scattering mechanisms. The first term is the contribution of the longitudinal viscous stresses, which is relatively small when the second condition of (5.2) is satisfied. The second and third terms of expression (6.2) are determined by the non-uniformity of the profiles of the velocity of the liquid and of the concentration of impurity in the cross-section.[1,2] In view of the large value of the Prandtl diffusion number ($\nu/D \gg 1$) the third term in expression (6.2) is many times greater than the second, if the solubility of the impurity in the liquid is significant (the ratio h/H is not small). The condition for the quantity (6.2) to be small compared with the frequency gives the limitation on the applicability of the approximation of an ideal compressible medium.

We will consider the case when the coefficient of elasticity of the film (5.7) is mainly determined by the elasticity of its surface, $\varepsilon = \varepsilon'$. Corresponding to relations (5.8) and (6.2), we will write the limitation which it is necessary to impose on the small value kh in order to satisfy the condition for the attenuation of the sound to be weak:

$$\frac{d}{\omega_0} = kh\left\{\frac{1}{\Delta^{1/2}} + \frac{\Delta^{1/2}}{12}\left(1 + \frac{\Pr}{W}\right)\right\} \ll 1, \quad \Delta = \frac{\varepsilon' h}{2\rho\nu^2} \tag{6.3}$$

It can be seen that when the coefficient of elasticity ε' of the surface changes, the energy losses per oscillation have a minimum. At the minimum point, the two-dimensional medium – the model of the film – is closest to an ideal fluid, and

$$\min\{d/\omega_0\} = 2kh\Delta_m^{-1/2}, \quad \Delta_m = 12/(1 + \Pr/W) \tag{6.4}$$

From expressions (6.4) we obtain the condition for a range of values of the coefficient of elasticity to exist, in which the sound waves are only slightly attenuated:

$$k^2h^2(1 + \Pr/W) \ll 3 \tag{6.5}$$

For the least value of the Prandtl number $\Pr \approx 10^3$, corresponding to low-viscosity liquids, and also for values of the ratio h/H that are not small, inequality (6.5) gives a considerable limitation of the wavelength with low attenuation for any values of the surface elasticity: $\lambda \gg 100h$.

Condition (6.5) also limits the applicability of the gas-dynamic approximation to a description of flows in a film with a soluble impurity. It is important that, if the impurity is insoluble, there is no similar limitation and the condition for a range of parameters to exist where there are weakly-attenuating sound waves will be the usual one: $kh \ll 1$. This indicates the qualitative difference in the dynamic behaviour of films with insoluble and soluble impurity.

Note that when the impurity is soluble, when $h^2 \gg D\tau$, the attenuation of small perturbations controls the parameter $D\tau/H^2$. When its values are not small, the attenuation of a sound wave is high and the gas-dynamic approximation is unsuitable.

For the limiting case $h^2 \ll D\tau$, we can propose a non-linear model of the motions of a film as a non-viscous compressible medium based on Eq. (5.4) with a pressure $\tilde{p}$, expressed in terms of σ as given by the first relation of (5.3). In this case we define the quantity σ using integral (4.5) taking into account the non-equilibrium and relations (1.5). Then, instead of the equilibrium pressure (5.3) we have the non-equilibrium pressure

$$\tilde{p} = \tilde{p}\left(h, \frac{dh}{dt}, \xi_i, T\right) \tag{6.6}$$

According to relation (6.6), the equation of state of a two-dimensional non-viscous compressible medium – the model of the film, includes the derivative with respect to time of the density of this medium. The presence of dh/dt in the equation of state corresponds to the fact that, during the flow of a two-dimensional medium energy dissipation occurs, due to the non-equilibrium nature of the impurity concentration profile in the cross section of the film.

7. Inertia effects in film dynamics

We will consider, corresponding to the profile of the velocity $\boldsymbol{v}'$ (1.1), the difference of the mean acceleration of the liquid from the acceleration defined by the mean velocity of the liquid in the cross-section

$$\overline{\frac{d\mathbf{v}'}{dt}} - \frac{d\bar{\mathbf{v}}'}{dt} = \frac{1}{5}(\mathbf{u}\nabla)\mathbf{u} \tag{7.1}$$

We will estimate $|(\mathbf{u}\nabla)\mathbf{u}|$, taking the smallness of the quantity $h^2/(\nu\tau) \ll 1$ into account.

For motion of the film as a two-dimensional compressible medium, we obtain from Eqs. (1.2) and (1.3) that the second velocity $u \sim h^2(\nu\tau)^{-1}v^*$, where v^* is the characteristic change in the velocity v. Hence it follows that the right-hand side of Eq. (7.1) is of the order of the square of the small parameter.

For slow motions, when the velocities can be of one order, $u \sim v^*$, it is more convenient to use the pressure p inside the film in estimates. We can then write

$$|(\mathbf{u}\nabla)\mathbf{u}| \sim h^2(\nu\tau)^{-1}|\nabla p|$$

Hence it follows that the right-hand side of equality (7.1) makes an asymptotically small contribution to the momentum equation (1.2), which can be neglected in the framework of the model of quasi-steady flow in the film considered.

8. The equations of motion of films for small Mach numbers

We will consider the limit of low compressibility of the surface (high elasticity ε') and derive the equations of motion of the film for small Mach numbers, also obtaining other limitations. We will denote by v^* the characteristic value (scale) of the velocities u and v, for motions of finite amplitude $v^*\tau = \lambda$. It is necessary to obtain the conditions for which the condition of low compressibility holds for the velocity field $\boldsymbol{v}$:

$$\operatorname{div}\mathbf{v} \ll v^*\lambda^{-1} \tag{8.1}$$

To solve this problem we reduce the equations of motion to dimensionless form for specified scales of the quantities. We will consider arbitrary smooth solutions of the equations.

For the case of an insoluble impurity, we obtain from Eq. (1.6) the condition for which condition (8.1) is satisfied,

$$\Gamma_1^* \ll \Gamma_0, \quad \Gamma_1 = \Gamma - \Gamma_0 \tag{8.2}$$

This indicates that the change in the surface concentration should be small.

For the case of a soluble impurity, we will use condition (8.2) in order to linearize the adsorption equation in the small neighbourhood of $c = c_0$. Then, from an analysis of the solutions of the problem for the equations of impurity transfer with linear conditions on the surface, we obtain two limitations, which are necessary to satisfy condition (8.1):

$$\left(1 + \frac{\sqrt{D\tau}}{H}\right)\frac{\Gamma_1^*}{\Gamma_0} \ll 1, \quad h \geq 2\sqrt{D\tau}; \quad \left(1 + \frac{h}{2H}\right)\frac{\Gamma_1^*}{\Gamma_0} \ll 1,$$

$$h \leq 2\sqrt{D\tau}; \quad H = H(c_0) \tag{8.3}$$

For motions of small amplitude, when the change h_1 of the thickness is small, the small factor h_1/h_0 will occur on the right-hand sides of relations (8.2) and (8.3). It also occurs in the estimates of the scale Γ_1^* derived below, with the exception of the third (Π_3). We will consider finite changes in the thickness $h_1 \sim h_0$.

From the momentum Eq. (1.2) we obtain an expression for the scale Γ_1^* of the change in the surface concentration

$$\frac{\Gamma_1^*}{\Gamma_0} = \left(1+\frac{h}{2H}\right)^{-1}\max\{\Pi_1, \ldots, \Pi_6\} \tag{8.4}$$

where

$$\Pi_1 = M^2, \quad \Pi_2 = \left(\frac{\lambda}{\tau c_s}\right)^2, \quad \Pi_3 = \frac{g\lambda}{c_s^2},$$

$$\Pi_4 = \frac{\sigma h^2}{4\varepsilon'\lambda^2}, \quad \Pi_5 = \frac{hp_e}{2\varepsilon'}, \quad \Pi_6 = \frac{\mu h}{2\varepsilon'\tau}$$

$$\left(M = \frac{\upsilon^*}{c_s}, \quad c_s^2 = \frac{2\varepsilon'}{\rho h}\right) \tag{8.5}$$

From relations (8.2)–(8.4) we obtain the conditions for the limit (8.1) of low compressibility of the film surface to be satisfied

$$\Pi_\beta \ll 1, \quad R = \frac{H+h/2}{H+\sqrt{D\tau}} < 1; \quad \Pi_\beta \ll R, \quad R \ge 1; \quad \beta = 1, \ldots, 6 \tag{8.6}$$

The second condition of (8.6) differs from the first only for $h \gg H, \sqrt{D\tau}$, when $R \gg 1$. The parameter Π_1 is the square of the Mach number, Π_2, Π_3 and Π_4 take into account the characteristic frequency for small perturbations, the mass force the Laplace pressure, respectively the parameter Π_5 is important in the case of super-thin films, and Π_6 takes into account the viscous stresses in the longitudinal flow. The factor h_0/h_1 occurs in the parameter Π_3 for small changes in the thickness.

The conditions $\Pi_4, \Pi_5, \Pi_6 \ll 1$ have analogies in the case of an ideal compressible medium in the form of conditions (5.9)–(5.11).

The fifth condition of incompressibility of the surface $\Pi_5 \ll 1$ corresponds to the effect of stabilization of a super-thin film[2] due to the action of the elasticity ε'. The stabilization of an unstable film consists of a multiple reduction in the rate of increase of small perturbations, ensured due to the transition of the instability of the film from the high-frequency branch of the solution of the dispersion equation to its low-frequency branch (the imaginary value of the velocity of sound changes into a real value). For conditions (8.5) and (8.6) the equations of motion (1.2)–(1.7) are simplified:

$$h\rho\frac{d\upsilon_i'}{dt} = 2\nabla_i\sigma + h\rho g_i + h\nabla_i\left(\frac{\sigma}{2}\nabla^2 h - p_e\right) + \nabla_j\{h\mu(\nabla_i\upsilon_j + \nabla_j\upsilon_i)\}$$

$$\boldsymbol{\upsilon}' = \mathbf{u} + \boldsymbol{\upsilon}, \quad \mathbf{u} = -\frac{h}{6\mu}\nabla\sigma, \quad \operatorname{div}\boldsymbol{\upsilon} = 0, \quad \frac{\partial h}{\partial t} + \operatorname{div}(h\boldsymbol{\upsilon}') = 0 \tag{8.7}$$

We have the equations of the flow of a two-dimensional fluid with density ρh and two velocities $\boldsymbol{\upsilon}$ and $\mathbf{u}$, where the incompressibility equation is satisfied for the first. It is important that the system of Eq. (8.7) does not contain the dependence of the surface tension on the impurity concentration, and the impurity transfer and surface adsorption equations. The effect of the corresponding parameters was retained in limitations (8.6), which must be verified when formulating and solving problems of film dynamics.

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