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QUASI-STEADY-STATE FLOWS IN A LIQUID FILM IN A GAS: COMPARISON OF TWO METHODS OF DESCRIBING WAVES

O. V. Voinov

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The motion of thin films of a viscous incompressible liquid in a gas under the action of capillary forces is studied. The surface tension depends on the surfactant concentration, and the liquid is nonvolatile. The motion is described by the well-known model of quasi-steady-state viscous film flow. The linear-wave solutions are compared with the solution using the Navier–Stokes equations. Situations are studied where a solution close to the inviscid two-dimensional solutions exists and in the case of long wavelength, the occurrence of sound waves in the film due to the Gibbs surface elasticity is possible. The behavior of the exact solutions near the region of applicability of asymptotic equations is studied, and nonmonotonic dependences of the wave characteristics on wavenumber are obtained.

Key words: *viscous incompressible liquid, capillary forces, surface tension, film in a gas, wave dynamics.*

1. Model of Quasi-Steady-State Flows in a Film. The thermodynamic foundations of the dynamics of liquid films with variable surface tension were developed in Gibbs papers on interfacial thermodynamics [1]. Gibbs performed important studies of the dynamics of liquid films with a surfactant, which were continued by Mysels et al. [2]. The motion of liquid films in a gas were theoretically and experimentally studied by Taylor [3]. Taylor's theory of the dynamics of free liquid films and the theory of shallow water on a horizontal surface [4] are based on the ideal fluid model. Unlike in the problems considered in these theories, in the case where the film surface is subjected to tangential stress, liquid viscosity plays an important role [5, 6]. Due to viscosity, the velocity of the liquid can change insignificantly over the film thickness. In a free film, sound waves due to the Gibbs surface elasticity [5] are possible.

We consider a film symmetric about the plane $x_3 = 0$ using the following assumptions. The distance λ at which the flow parameters vary is large compared to the film thickness h ($\lambda \gg h$). The characteristic time τ far exceeds the time of the transfer vortex across the film: $h^2 \ll \nu\tau$ (ν is the kinematic viscosity). This condition is assumed to be satisfied for the time τ calculated from the value of λ and the characteristic liquid velocity: $\tau = \lambda/v^*$, which is equivalent to the smallness of the reduced Reynolds number of the hydrodynamic theory of lubrication: $v * h^2/(\lambda\nu) \ll 1$. Under these assumptions [6], the flow at each cross-sections of the film is close to steady-state flow with quadratic velocity profile and coefficients that vary slowly over the coordinates x_1, x_2:

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

Here v_i is the velocity of the film surface and u_i is the average flow velocity relative to the surface. In the equations of motion of the film, the acceleration of the liquid is determined primarily by the velocity $\boldsymbol{v}$ [6]. To take into account the effect of the velocity $\boldsymbol{u}$ on the small attenuation of waves in the film [7], we find the acceleration of the liquid from the average value of the velocity $\boldsymbol{v}'$ in the film. The difference between the average acceleration of

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Tyumen' Department of the Khristianovich Institute of Theoretical and Applied Mechanics, Siberian Division, Russian Academy of Science, Tyumen' 625000; o.v.voinov@mtu-net.ru. Translated from Prikladnaya Mekhanika i Tekhnicheskaya Fizika, Vol. 48, No. 3, pp. 103–111, May–June, 2007. Original article submitted September 28, 2006.

the liquid in the film, determined by relation (1.1), and the acceleration calculated from the average velocity $\overline{\boldsymbol{v}'}$, is proportional to the small quantity $(\boldsymbol{u}\cdot\nabla)\boldsymbol{u}$, whose value is insignificant in the adopted film flow model.

The equations of quasi-steady-state film flows [6] are written as

$$h\rho\,\frac{d\overline{v_i'}}{dt}=2\nabla_i\sigma+h\rho g_i+h\nabla_i\Big(\frac{\sigma}{2}\,\nabla^2h-p_e\Big)+\nabla_j\{h\mu(2\delta_{ij}\operatorname{div}\boldsymbol{v}+\nabla_iv_j+\nabla_jv_i)\};\tag{1.2}$$

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

$$\frac{\partial h}{\partial t}+\nabla\cdot h\overline{\boldsymbol{v}'}=0,\qquad \overline{\boldsymbol{v}'}=\boldsymbol{u}+\boldsymbol{v};\tag{1.4}$$

$$\sigma=\sigma(\Gamma,T),\qquad \Gamma=\Gamma(c,T),\qquad x_3=\pm h/2;\tag{1.5}$$

$$\frac{\partial\Gamma}{\partial t}+\operatorname{div}(\Gamma\boldsymbol{v})=-D\,\frac{\partial c}{\partial x_3}\Big(x_1,x_2,\frac{h}{2},t\Big);\tag{1.6}$$

$$\frac{\partial c}{\partial t}+v_j'\nabla_jc-(x_3\operatorname{div}\boldsymbol{v})\nabla_3c=D\nabla^2c,\qquad x_3\in\Big(-\frac{h}{2},\frac{h}{2}\Big).\tag{1.7}$$

Here δ_{ij} is the Kronecker symbol, the summation is performed over the repeated index $j=1$, 2, g_i is the body force, ρ is the density, μ is the dynamic viscosity, D is the diffusion coefficient, c and Γ are the volumetric and surface impurity concentrations, whose profiles are symmetric about the point $x_3=0$; the concentration Γ is related to the value of c on the surface by the adsorption equation (1.5); the surface tension σ depends on Γ according to the first equation in (1.5); the gas temperature T and pressure are constant; p_e is an additional term in the pressure expression for ultrathin films (differs in sign from the wedging pressure). The expression for the effective pressure p_e takes into account, in particular, the action of Van der Waals forces, which is described by similar equations for a film in a gas and a film on a solid wall [5, 8–13]. Equations (1.6) and (1.7) describe impurity transfer in the liquid. The possible effect of the surface rheology [6] is not considered.

There is divergent form of the momentum equation (1.2), which is suitable for writing the integrals of the equations [14]:

$$\frac{\partial}{\partial t}\,(h\rho v_i')+\nabla_jI_{ij}=h\rho g_i,\qquad I_{ij}=\rho hv_i'v_j'-2s_{ij}-hp_{ij}-E\delta_{ij},$$

$$s_{ij}=\sigma(1+|\nabla z|^2/2)\delta_{ij}-\sigma(\nabla_iz)\nabla_jz,\qquad z=h/2,$$

$$p_{ij}=\delta_{ij}((\sigma/2)\nabla^2h-p_e)+\mu(2\delta_{ij}\operatorname{div}\boldsymbol{v}+\nabla_iv_j+\nabla_jv_i).$$

Here v_i' is the average velocity and E is the primitive of $p_e=\partial E/\partial h$.

In the case of an insoluble impurity in a liquid, Eqs. (1.5)–(1.7) reduce to the two equations

$$\sigma=\sigma(\Gamma,T),\qquad \frac{\partial\Gamma}{\partial t}+\operatorname{div}(\Gamma\boldsymbol{v})=0.\tag{1.8}$$

Using (1.8), we write the surface elasticity coefficient as

$$\varepsilon'=-\Gamma\,\frac{\partial\sigma}{\partial\Gamma}.\tag{1.9}$$

In some cases, the solution of the problem for an insoluble impurity can be used as an approximate solution of the problem for a soluble impurity.

First of all, it is known under what conditions the impurity solubility effects are insignificant. We consider small perturbations of the film equilibrium. If the film thickness is small enough [8, 14]:

$$h\ll H,\qquad H=\partial\Gamma/\partial c$$

a soluble impurity behaves as an insoluble one and the transfer equation (1.8) has a small error: $h/H\ll1$ (c is the concentration on the surface). The diffusion of the impurity affects sound waves in the film [14], increasing their attenuation. A decrease of h/H leads to a reduction in the diffusion effect.

In addition, there is an analogy between the behavior of films with soluble and insoluble impurities [8, 14]. Let the average velocity in a film be close to the velocity of the surface $\boldsymbol{v}$, i.e., $u \approx 0$, and let the inequalities $h^2 \ll D\tau \ll \lambda^2$ be satisfied. Then, $2\Gamma/h + c = f(\xi_i)$ (f is an arbitrary function of Lagrangian coordinates ξ_i); in this case, the surface tension is $\sigma = \sigma(h, \xi_i, T)$. Similarly to (1.9), the surface elasticity coefficient of the film is equal to

$$\varepsilon' = -\Gamma \frac{\partial \sigma}{\partial \Gamma} \Big(1 + \frac{h}{2H}\Big)^{-1}.$$

This analogy between the behavior of films with soluble and insoluble impurities can be valid in the case of sound waves [5], where the characteristic velocity values differ significantly ($u \ll v$), and it is absent for slow aperiodic motions with a low velocity of the surface $\boldsymbol{v}$ (the most interesting case for such motions). The existence of the analogy is also limited by the condition $kh \ll \mathrm{Pr}^{-1/2}$ ($\mathrm{Pr} = \nu/D$ is the diffusion Prandtl number, which is large for liquids [14]).

Below, the case of an insoluble impurity is considered.

2. Problem of Waves within the Framework of Linearized Navier–Stokes Equations. We consider the case of small perturbations periodic in the x axis, where the film thickness is given by the relation

$$h = h_0 + h_1 \exp(\alpha' t - ikx). \tag{2.1}$$

The linear problem of waves in a liquid with a surfactant is considered in [15] for a liquid of infinite depth. This problem differs significantly from the problem of a film in which the occurrence of sound waves [5] is possible.

For the solution of the problem of symmetric waves of small amplitude in the presence of an insoluble impurity [5], the following dispersion equation holds:

$$(\Omega \tanh a + (2 + \alpha^2)) \tanh(la) - 4l \tanh a + \delta(l - \Omega(\tanh(la) - l \tanh a)\alpha^{-2}) = 0; \tag{2.2}$$

$$l = \sqrt{\alpha + 1}, \quad a = \frac{1}{2} kh, \quad \alpha = \frac{\alpha'}{\nu k^2}, \quad \Omega = \frac{\sigma - 2Qk^{-2}}{\rho \nu^2 k}, \quad \delta = \frac{\varepsilon'}{\rho \nu^2 k}. \tag{2.3}$$

Here $Q = -\partial p_e/\partial h$. According to relations (2.2) and (2.3), the dependence of the exponent α' on the wavenumber k includes three parameters, two of which are incorporated in the parameter Ω.

3. Waves in a Film on the Basis of Asymptotic Equations of Motion. The equations of motion for the case of an insoluble impurity (1.1)–(1.4) and (1.8), with the use of relations (2.1), imply the equation

$$\alpha^2 + \alpha\Big(4 + \frac{1}{3}\delta a\Big) + \Omega a + \frac{\delta}{a} + \frac{a^2}{3\alpha}\delta\Omega = 0. \tag{3.1}$$

The dimensionless parameters (2.3) are written as

$$\delta = \Delta/a, \qquad \Omega = \Omega_\sigma a^{-1} + \Omega_e a^{-3},$$

where the elasticity coefficient Δ, the capillary pressure parameters Ω_σ, and the effective pressure Ω_e do not depend on the wavenumber k.

In the case of an significant effect of the liquid inertia during motion of the film (for example, in the case of sound waves), the modulus of the dimensionless parameter is significant: $|\alpha| \gg 1$. In this case, Eq. (3.1) has the same constraint as the model of quasi-steady-state flow: $h^2 \ll \nu\tau$. In view of relations $|\alpha'| \approx 1/\tau$, this implies

$$|\alpha| a^2 \ll 1. \tag{3.2}$$

Inequality (3.2) is valid when on the film surface is subjected to tangential stress due to the elasticity coefficient $\varepsilon' \neq 0$ ($\delta \neq 0$), and it is not extended to problems with no elasticity.

4. Relationship between the Dispersion Equations of the Approximate and Exact Theories. We consider the dispersion equation (3.1) of the approximate theory and the asymptotic form (for $a \ll 1$) of the dispersion equation (2.2) of the linear-wave problem within the framework of the Navier–Stokes equations. We take into account the main terms of the Taylor expansions for $a = 0$ for the functions in (2.2). We omit the small terms $O(a^2)$ but retain the small terms $O(\alpha a^2)$. As a result, the limiting form of the exact equation (2.2) for long waves coincides with the approximate equation (3.1), which follows from the asymptotic equations.

For a considerable kh, the approximate values of the exponent $\alpha(k)$ differ from the exact values continuing the solution for small kh. To determine the region of applicability of the approximate solution, we consider it up to values $kh \approx 1$, together with the exact solution.

5. Roots of the Dispersion Equation. We consider the solution for a relatively small effect of the liquid viscosity: $\lambda^2 \gg \nu\tau$ or $|\alpha| \gg 1$. We find two roots of Eq. (3.1) approximately for a small value of the number a, assuming, in addition, that $|\alpha| \gg 1$:

$$\alpha = \pm i\omega_0, \qquad \omega_0 = (\Omega a + \delta/a)^{1/2}. \tag{5.1}$$

In view of (3.2), this is true for

$$1 \ll \omega_0 \ll a^{-2}.$$

Here the second inequality is significant when $\delta \neq 0$. We note that solution (5.1) t corresponds to a small second velocity: $u \ll v$.

We write the high-frequency root of Eq. (3.1) in approximate form, assuming that the contribution of the capillary pressure parameter Ω_σ is small:

$$\alpha = \pm i a^{-1}(\Delta + \Omega_e)^{1/2}.$$

For $\Delta + \Omega_e < 0$, this implies exponential instability of the film [5]. If inequality $\Delta + \Omega_e > 0$ is valid, the dimensional frequency ω_0' is proportional to the wavenumber k [5], which corresponds to a sound wave. For $\Delta \gg \Omega_e$, we express the frequency in terms of the sound velocity c_s:

$$\omega_0' = c_s k, \qquad c_s = (2\varepsilon'/(\rho h))^{1/2}.$$

The dimensionless attenuation decrement β depends on the elasticity [7]:

$$\alpha = \pm i\omega_0 - \beta, \qquad \beta = 2 + \frac{1}{6}\,\frac{\Delta^2}{\Delta + \Omega a^3}. \tag{5.2}$$

Let us consider wave attenuation for one oscillation versus surface elasticity for a value of a small enough so that the inequality $\Omega a^3 \ll \Delta$ is satisfied. From (5.1) and (5.2) it follows that there is a minimum of the energy loss for one oscillation: $\min(\beta/\omega_0)$ at $\Delta = 12$. In this case, the wave is the closest to the wave in an ideal compressible medium.

We specify the root (5.1) of Eq. (3.1) by means of iteration:

$$\alpha = \pm i\omega_1 - \beta, \qquad \omega_1^2 = \omega_0^2 - \beta^2 + \frac{2}{3}\,\beta\,\frac{\Omega a \Delta}{\omega_0^2}. \tag{5.3}$$

The third root of Eq. (3.1) corresponds to possible aperiodic motions. In the presence of a root with a high frequency ($\omega_0 \gg 1$) and $a \ll 1$,

$$\alpha_3 \approx \alpha_{30} = -\frac{a^2}{3}\,\frac{\Omega\delta}{\omega_0^2}. \tag{5.4}$$

We specify the root (5.4) of Eq. (3.1) by means of iteration:

$$\alpha_3 = -(1/3)\Omega a \Delta(\omega_0^2 + \alpha_{30}^2 + \alpha_{30}(4 + \Delta/3))^{-1}. \tag{5.5}$$

We show that the third root is small compared to the dimensionless frequency ω_0. The elasticity coefficient $\delta \sim \Omega a^2$ is estimated as

$$\alpha_3 \sim a^2\omega_0^2 \ll \omega_0.$$

For a large value of the coefficient $\delta \gg \Omega a^2$, we have the estimate

$$\alpha_3 = -\Omega a^3/3 \ll a^2\omega_0^2 \ll \omega_0.$$

The small value of the root α_3 is responsible for the stabilization of the unstable film (for $\Omega_e < 0$) due to the impurity effect [5]. As the elasticity increases, the instability passes from the high-frequency branch of the solution of the dispersion equation to the branch of aperiodic motion with the long-wave perturbation growth increment decreasing many times.

6. Relations between the Wave Parameters $\alpha(k)$ of the Approximate and Exact Theories. We consider the dependences of the wave characteristics on the wavenumber for the ratio of the capillary pressure parameter to the elasticity coefficient $\Omega_\sigma/\Delta = \sigma/\varepsilon' = 10$. The special properties of ultrathin films are not considered: $\Omega_e = 0$.

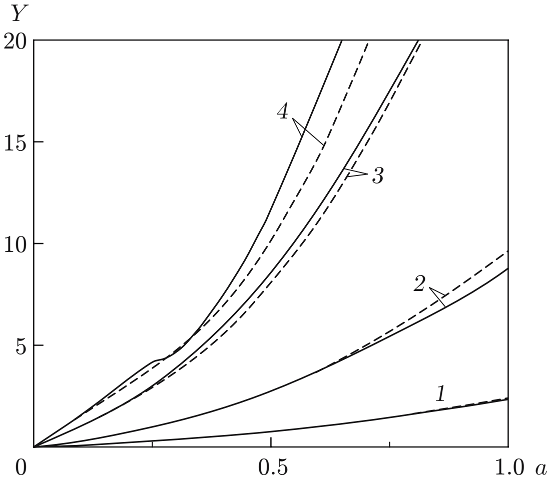

Fig. 1. Frequency versus wavenumber for $\Omega_\sigma = 10$ (1), 100 (2), 1000 (3), and 2285 (4): the solid curves are solutions of the exact equation (2.2); the dashed curves are calculated by formulas (5.2) and (5.3).

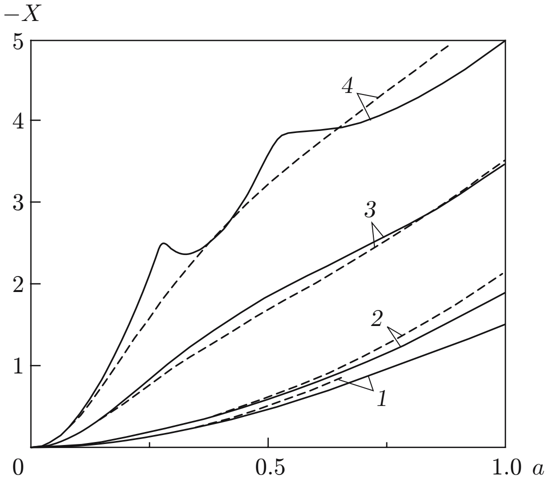

Fig. 2

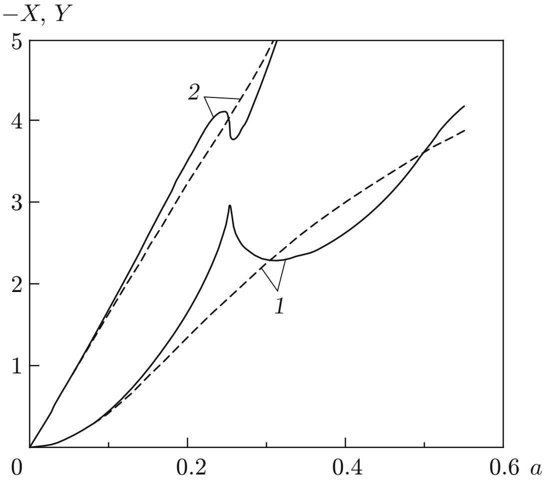

Fig. 3

Fig. 2. Attenuation decrement versus wavenumber (notation the same as in Fig. 1).

Fig. 3. Nonmonotonic curves of the attenuation decrement (1) and frequencies (2) versus wavenumber: the solid curves are solutions of the exact equation (2.2); the dashed curves are calculated by formulas (5.2) and (5.3).

We denote $\alpha a^2 = X + iY$. Figure 1 shows curves of the dimensionless frequency Y versus dimensionless wavenumber $a = kh/2$. The roots of Eq. (2.2) that at $a \to 0$ becomes roots of Eq. (3.1) are considered. As the elasticity coefficient Δ increases, the region in which the exact and asymptotic theories agree becomes narrower. The solid curve 4 has the point of inflection $a = 0.2772$. This implies that with a further increase in the parameter Δ, the dependence $Y(a)$ for $a < 1$ is nonmonotonic.

Figure 2 shows curves of the dimensionless attenuation decrement $-X$ versus wavenumber a. The exact curve 4 is nonmonotonic. Nonmonotonicity appears for $\Delta = 209.69$, where there is the point of inflection corresponding to the value $a = 0.3169$. The nonmonotonicity of the dependences of the frequency and attenuation decrement on wavenumber is not described by asymptotic theory.

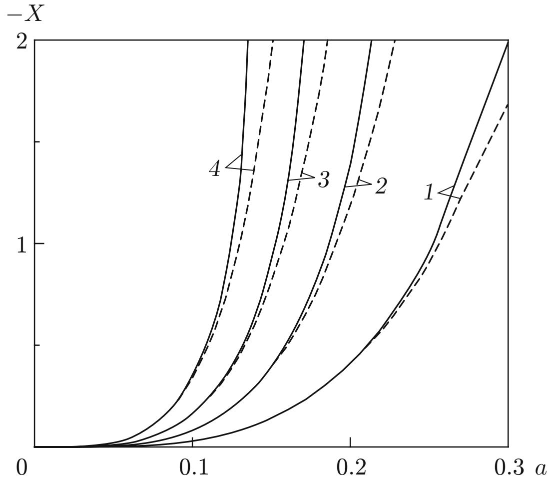

Fig. 4

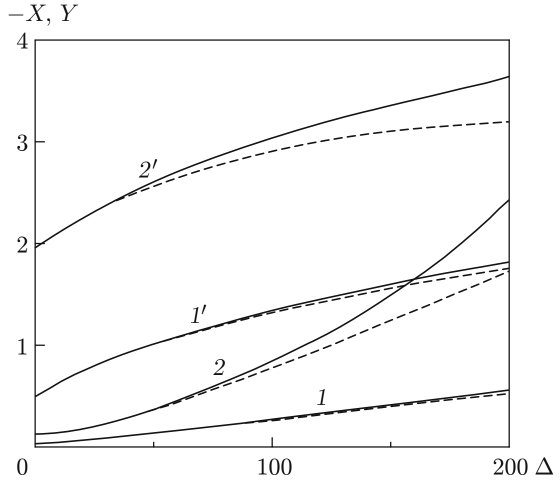

Fig. 5

Fig. 4. Attenuation decrement of aperiodic motion for $\Omega_\sigma = 1000$ (1), 2600 (2), 5000 (3), and 10^4 (4): the solid curves are solutions of the exact equation (2.2); the dashed curves are calculated by formula (5.5).

Fig. 5. Attenuation decrement (1 and 2) and frequencies ($1'$ and $2'$) versus elasticity coefficient for $\Omega_\sigma = 1000$: the solid curves are solutions of the exact equation (2.2); the dashed curve are calculated by formulas (5.2) and (5.3); curves 1 and $1'$ refer to $a = 0.125$ and curves 2 and $2'$ refer to $a = 0.25$.

The point $a = 0$ corresponds to the absolute minimum of the oscillation attenuation, near which the energy loss due to oscillations disappear: $\beta/\omega \to 0$ as $a \to 0$.

Figure 3 gives nonmonotonic dependences of the attenuation decrement and frequency on the wavenumber. According to the exact solution, the attenuation decrement is maximal at $a = 0.2544$. At first sight, the solid curve 1 has a corner point. However, calculations with small a step in the wavenumber a show that this curve is smooth. For small a, the exact and asymptotic solutions coincide.

For a small value of a, all dependences of the frequency on the wavenumber in Figs. 1 and 3 are nearly linear. The linear asymptotics corresponds to a sound wave in a film [5] with a small attenuation decrement (of order a^2).

Equation (3.1) has a real third root α_3 which corresponds to aperiodic motion. The dependences of the coefficient $X = \alpha_3 a^2$ on the wavenumber for various values of Ω_σ are given in Fig. 4. The regions of applicability of the approximate solutions correspond to values $|X| < 1$. It is evident that the asymptotic solution agrees well with the exact solution. From Fig. 4, it follows that purely attenuating motions with a significant effect of the inertia of the liquid are possible.

The effect of the elasticity coefficient Δ on the wave frequency and attenuation decrement for $\Omega_\sigma = 1000$ is shown in Fig. 5. It is evident that the exact solution agrees well with the linear asymptotics of the attenuation decrement $-X(\Delta)$ (curves 1 and 2). The asymptotics takes into account the effect of the second velocity u on the oscillation attenuation [7]. As the elasticity increases, the attenuation decrement $|X|$ changes many times compared to the value for $\Delta = 0$ determined by viscous stresses in the longitudinal flow (curves 1). The effect of the elasticity on the attenuation [7, 14] is determined by the small value of the ratio $h^2/(\nu\tau)$ where $\tau = 1/\omega_0'$ (ω_0' is the dimensional frequency).

In the small-wavenumber limit ($kh \to 0$) for the solutions describing sound waves [5], neither viscosity nor the second velocity u are of significance in the momentum equation (1.2). In this case, the film is similar to an ideal compressible medium if the elasticity coefficient Δ is bounded. In the case $\Delta \approx a^{-2}$, the wave attenuation is comparable to the frequency and asymptotic theory is inapplicable since condition (3.2) is not satisfied. We note that inequality (3.2) is used to derive an approximate solution of the dispersion equation taking into account the surface elasticity [5].

Figure 6 shows curves of the dimensionless parameter $X = \alpha_3 a^2$ versus elasticity coefficient Δ for $\Omega_\sigma = 1000$ for the case of aperiodic film motion. For $a = 0.125$, the exact values and solution (5.5) differ by less than 1% (curves 1). In this case, X depends appreciably on the elasticity coefficient Δ in the range of small values of the

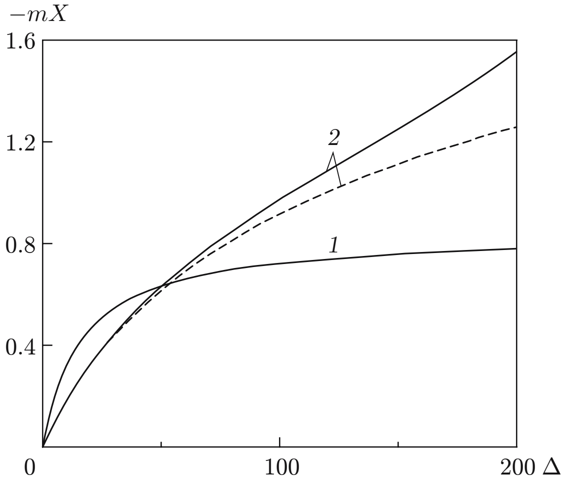

Fig. 6. Attenuation decrement of aperiodic motions versus elasticity coefficient: the solid curves are solutions of the exact equation (2.2); the dashed curves are calculated by formula (5.5); curve 1 refers to $m = 10$ and $a = 0.125$; curves 2 refer to $m = 1$ and $a = 0.25$.

latter, and in the range $20 < \Delta < 200$, the value of X varies only slightly. This region of slight variation of X is absent for $a = 0.25$. The approximate solutions are suitable for small a and $|X| < 1$.

7. Conclusions. The above comparison of two methods of describing linear waves in a liquid film in the presence of an insoluble surfactant leads to the following conclusions.

Nonmonotonic dependences of the wave frequency and the attenuation decrement on the wavenumber outside the region of applicability of the asymptotics are possible.

For small values of the wavenumber, the region of applicability of the approximate solutions is adequately estimated by the equality for which the strong inequality (3.2) is not satisfied.

The slope of the curve of frequency versus wavenumber at the coordinate origin corresponds to the sound velocity in the film, and the asymptotic model [6] and the exact theory of linear waves [5] agree.

Aperiodic film motion is possible for a great effect of not only viscosity but also inertia forces.

For a certain value of the surface elasticity coefficient in the long wavelength limit, the energy loss for the period become minimal and the ratio of the sound velocity in the film to the corresponding value in an ideal two-dimensional compressible medium is the closest to unity.

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