DYNAMICS OF FREE LIQUID FILMS

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1. We will term a layer of viscous incompressible liquid moving in another liquid or gas a free film if its thickness h measured along the normal n to the mean surface Γ is small in comparison to the characteristic scale of motion while the gradients of velocity v and temperature θ within the layer are finite as $h \to 0$. The latter will be true if the viscosity and thermal conductivity coefficients of the external liquid do not exceed the corresponding characteristics of the film material. Then, in the first approximation, the position of the film may be specified by a two-dimensional surface Γ and its dynamics may be described by parameters distributed over Γ, assuming v and θ to be continuous functions of the point in space x and time t everywhere (local thermodynamic equilibrium principle). In particular, Lagrangian coordinates for the film and enveloping phase can be defined, permitting use of a phenomenological approach to derivation of the equations of motion of Γ [1]. The basic principles of thermodynamics and rheological relationships lead to a closed problem in dynamics of thin liquid films, containing within itself at h = 0 the problem of thermocapillary convection [2], while the thermodynamic relationships transform to the classical Gibbs conditions on an interphase boundary. The equations of motion of isothermal free films of viscous and elastoviscous liquids were derived in [3].

We introduce the notation G for the metric (unit) space tensor, whereupon $G_\Gamma = G - \mathbf{nn}$ (with the vector multiplication being tensor) is the metric tensor of the surface Γ with the aid of which we project on Γ the tensors and differential operators. For example, the spatial gradient ∇ is transformed to a surface gradient $\nabla_\Gamma = G_\Gamma \cdot \nabla$ (the dot denoting the inner product), while the volume deformation rate tensor $D = (\nabla v)_{\mathrm{sym}}$ generates a surface deformation rate tensor $D_\Gamma = G_\Gamma \cdot D \cdot G_\Gamma = (\nabla_\Gamma v \cdot G_\Gamma)_{\mathrm{sym}}$. Since the spur D_Γ is equal to the surface divergence of the total velocity vector, for the material area element J_Γ of the surface Γ we have the expression $\dot{J}_\Gamma = J_\Gamma \operatorname{div}_\Gamma v$ (for the material volume element J we have $\dot{J} = J \operatorname{div} v$). Here and below, a dot above indicates the full time derivative in the particle. We note that $\operatorname{div}_\Gamma v = \operatorname{div}_\Gamma(G_\Gamma \cdot v) + k\, v \cdot n$ ($k = \operatorname{div}_\Gamma n$ is the sum of the main curvatures of Γ). Also valid is the important expression $\operatorname{div}_\Gamma G_\Gamma = -k\mathbf{n}$. In fact, in a Euclidean space the tensor G is constant, while the curvature tensor $\nabla_\Gamma \mathbf{n}$ does not have normal components, so that $\operatorname{div}_\Gamma(G - \mathbf{nn}) = -\operatorname{div}_\Gamma(\mathbf{nn}) = -k\,\mathbf{n}$.

Let ρ, e, T, and $\mathbf{q}$ be the mass density, internal energy, stress tensor, and thermal flux vector of the volume phase; ρ_Γ, e_Γ, T_Γ, and $\mathbf{q}_\Gamma$ are the analogous characteristics of the film surface phase. Then the differential laws of conservation of mass, momentum, and energy take on the form [1]

$$\dot{\rho} + \rho \operatorname{div} \mathbf{v} = 0,\ \rho(\dot{\mathbf{v}} - \mathbf{g}) = \operatorname{div} T, \qquad \rho\dot{e} = T : \nabla \mathbf{v} - \operatorname{div} \mathbf{q} \text{ outside } \Gamma; \tag{1.1}$$

$$\dot{\rho}_\Gamma + \rho_\Gamma \operatorname{div}_\Gamma \mathbf{v} = 0,\ \rho_\Gamma(\dot{\mathbf{v}} - \mathbf{g}) = \operatorname{div}_\Gamma T_\Gamma + \mathbf{n}\cdot[T], \qquad \rho_\Gamma \dot{e}_\Gamma = T_\Gamma : \nabla_\Gamma \mathbf{v} - \operatorname{div}_\Gamma \mathbf{q}_\Gamma - \mathbf{n}\cdot[\mathbf{q}] \text{ on } \Gamma, \tag{1.2}$$

where g is the external mass force density; the colon denotes the scalar product (convolution of second range tensors, the square brackets correspond to the operation of calculating the change in a function upon traversing the surface Γ in the positive direction of the normal n, i.e.,

$$[f](\mathbf{x}) = \lim_{\eta \to 0} \operatorname{sgn} \eta \,\{f(\mathbf{x} + \eta\mathbf{n}) - f(\mathbf{x} - \eta\mathbf{n})\}, \quad \mathbf{x} \in \Gamma.$$

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Equations (1.1) and (1.2) are a consequence of the integral laws of conservation of mass, momentum, and energy for an arbitrary material volume (phase transitions are absent, i.e., the surface and volume phases may exchange momentum and energy, but not mass):

$$\frac{d}{dt}\int\limits_{\omega_t}\rho d^3x=0,\quad \frac{d}{dt}\int\limits_{\gamma_t}\rho_\Gamma d^2x=0,$$

$$\frac{d}{dt}\left\{\int\limits_{\omega_t}\rho\mathbf{v}d^3x+\int\limits_{\gamma_t}\rho_\Gamma\mathbf{v}d^2x\right\}=\int\limits_{\partial\omega_t}\boldsymbol{\nu}\cdot Td^2x+\int\limits_{\partial\gamma_t}\boldsymbol{\nu}_\Gamma\cdot T_\Gamma d^1x+\int\limits_{\omega_t}\rho\mathbf{g}d^3x+\int\limits_{\gamma_t}\rho_\Gamma\mathbf{g}d^2x,$$

$$\frac{d}{dt}\left\{\int\limits_{\omega_t}\rho\left(\frac{|\mathbf{v}|^2}{2}+e\right)d^3x+\int\limits_{\gamma_t}\rho_\Gamma\left(\frac{|\mathbf{v}|^2}{2}+e_\Gamma\right)d^2x\right\}=\int\limits_{\partial\omega_t}\boldsymbol{\nu}\cdot(T\cdot\mathbf{v}-\mathbf{q})\,d^2x+$$

$$+\int\limits_{\partial\gamma_t}\boldsymbol{\nu}_\Gamma\cdot(T_\Gamma\cdot\mathbf{v}-\mathbf{q}_\Gamma)\,d^1x+\int\limits_{\omega_t}\rho\mathbf{g}\cdot\mathbf{v}d^3x+\int\limits_{\gamma_t}\rho_\Gamma\mathbf{g}\cdot\mathbf{v}d^2x.$$

Here $\gamma_t = \Gamma \cap \omega_t$, $\boldsymbol{\nu}$ and $\boldsymbol{\nu}_\Gamma$ are unit external normals to the boundaries $\partial\omega_t$ and $\partial\gamma_t$, respectively, while $\boldsymbol{\nu}_\Gamma$ is tangent to the surface Γ.

The law of conservation of angular momentum is insured by the requirement of symmetry of the tensors T and T_Γ. Moreover, the condition of materiality of Γ and the local thermodynamic equilibrium principle lead to additional equalities

$$\dot{\mathbf{x}}=\mathbf{v},\ [\mathbf{v}]=\mathbf{0},\ [\theta]=0 \text{ on } \Gamma. \tag{1.3}$$

To complete Eqs. (1.1)-(1.3), we must turn to the film thermodynamics and rheology.

2. Initially we recall the defining relationships of the volume phase, which we will consider to be a viscous thermally conductive compressible gas. As is well known, the basic thermodynamic identity for the parameters ρ, e, p (pressure), θ (absolute temperature) is equivalent to the fact that the expression $(1/\theta)\{de + pd(1/\rho)\}$ is the complete differential of the entropy per unit mass. This latter reduces to the consistency condition $de \wedge d(1/\theta) = d(p/\theta) \wedge d(1/\rho)$, which for the functions $e(\rho, \theta)$ and $p(\rho, \theta)$ can also be written in the form

$$\rho^2\partial e/\partial\rho=-\theta^2\partial(p/\theta)/\partial\theta. \tag{2.1}$$

In particular, it follows from Eq. (2.1) that the energy of an ideal gas ($p = R\rho\theta$) depends solely on temperature. The classical Stokes and Fourier laws specify the rheology of the medium

$$T=(-p+\lambda\operatorname{div}\mathbf{v})G+2\mu D,\ \mathbf{q}=-\varkappa\nabla\theta, \tag{2.2}$$

where λ, μ, and κ are viscosity and thermal conductivity coefficients of the volume phase.

Analogous defining expressions can be written for the film, taking into account certain differences (as a continuous medium) from the volume phase. In fact, with respect to the external medium the film is not only a region of inhomogeneity, but also an anisotropic one, i.e., the stress tensor will not be spherical even for an equilibrium situation (the film itself is in tension, and the external medium in compression). In fact, disruption of Pascal's law occurs near the face interphase boundaries of the film. However, from a phenomenological viewpoint it is preferable to ascribe the anisotropy of the interphase boundaries to the film itself, with the phase interaction energy being referred to a unit mass of the film material.

Let τ be the thermodynamic stress of the film; p_*, the hydrodynamic pressure; ρ_*, the density of the film material; whereupon the basic thermodynamic identity leads to $(1/\theta)\cdot\{de_\Gamma + p_*d(1/\rho_*) - \tau d(1/\rho_\Gamma)\}$. Limiting ourselves to the case of incompressible film material ($\rho_* = \text{const}$) and taking $\varepsilon = \rho_\Gamma e_\Gamma$, we obtain an expression for the differential entropy $(1/\theta)\{d(\varepsilon/h) - \tau d(1/h)\}$, from which follows the consistency condition $d(1/\theta) \wedge d(\varepsilon/h) = d(\tau/\theta) \wedge d(1/h)$ (the equation which is asymptotically precise as $h \to 0$, $\rho_\Gamma = \rho_*h$ is valid). Thus, the functions $\varepsilon(h, \theta)$ and $\tau(h, \theta)$ related by the expression

$$h^2\partial(\varepsilon/h)/\partial h=\theta^2\partial(\tau/\theta)/\partial\theta, \tag{2.3}$$

completely define the film thermodynamics.

It is obvious that in equilibrium $T_\Gamma = \tau G_\Gamma - p_* hG$, where the pressure p_* at $\rho_* = \text{const}$ is no longer a thermodynamic quantity and must be defined from the condition of incompressibility of the film material. For stability of the film as a thermodynamic system the inequality $\partial\tau(h, \theta)/\partial h < 0$ must be satisfied, this following from the principle of minimum free energy. Because of the latter the limit of τ as $h \to 0$ is positive and in the first approximation $T_\Gamma \cdot \mathbf{n} = 0$.

We will impose on the nonequilibrium processes the condition of tangency to the surface Γ of external momentum and heat fluxes

$$T_\Gamma \cdot \mathbf{n} = \mathbf{0}, \quad \mathbf{q}_\Gamma \cdot \mathbf{n} = 0. \tag{2.4}$$

Then the axiom of isotropic linear dependence of T_Γ on D_Γ and of $\mathbf{q}_\Gamma$ on $\nabla_\Gamma\theta$ leads to analogs of the Stokes and Fourier laws, Eq. (2.2)

$$T_\Gamma = (\tau + \lambda_\Gamma \operatorname{div}_\Gamma \mathbf{v}) G_\Gamma + 2\mu_\Gamma D_\Gamma, \quad \mathbf{q}_\Gamma = -\varkappa_\Gamma \nabla_\Gamma \theta, \tag{2.5}$$

where λ_Γ, μ_Γ, and κ_Γ are viscosity and thermal conductivity coefficients of the surface phase.

We now transform Eq. (1.2) by introducing the heat capacity per unit film surface $\alpha(h, \theta) = \partial\varepsilon(h, \theta)/\partial\theta$ and the dissipative function $\Phi_\Gamma = \lambda_\Gamma(\operatorname{div}_\Gamma \mathbf{v})^2 + 2\mu_\Gamma D_\Gamma : D_\Gamma$ to a closed system of equations for h, v, and θ:

$$\begin{gathered} \dot{h} + h \operatorname{div}_\Gamma \mathbf{v} = 0, \quad \rho_* h (\dot{\mathbf{v}} - \mathbf{g}) = \operatorname{div}_\Gamma T_\Gamma + \mathbf{n} \cdot [T], \\ \alpha\dot{\theta} = \theta \frac{\partial\tau}{\partial\theta} \operatorname{div}_\Gamma \mathbf{v} + \Phi_\Gamma - \operatorname{div}_\Gamma \mathbf{q}_\Gamma - \mathbf{n} \cdot [\mathbf{q}] \text{ on } \Gamma. \end{gathered} \tag{2.6}$$

where thermodynamic condition (2.3) is used.

3. We will now find the relationship of λ_Γ, μ_Γ, and κ_Γ to the viscosity coefficient μ_* and the thermal conductivity κ_* of the film material. Let $T_* = -p_* G + 2\mu_* D_*$ be the stress tensor, D_* the deformation rate tensor, calculated from the velocity field $\mathbf{v}_*$ of the film material. From the noncompressibility condition we have the asymptote as $h \to 0$

$$\mathbf{v}_* = \mathbf{v} - z \operatorname{div}_\Gamma \mathbf{v}\mathbf{n}, \quad |z| < h/2,$$

where v is the velocity of the mean surface Γ, z is the coordinate measured along $\mathbf{n}$. It is obvious that

$$\begin{gathered} \mathbf{v} = \int\limits_{-h/2}^{h/2} \mathbf{v}_* dz, \quad T_\Gamma = \tau G_\Gamma + \int\limits_{-h/2}^{h/2} T_* dz = \\ = \tau G_\Gamma - p_* hG + 2\mu_* h (D_\Gamma - \operatorname{div}_\Gamma \mathbf{v}\mathbf{n}\mathbf{n}). \end{gathered}$$

Meanwhile, Eq. (2.4) leads to the equality $p_* = -2\mu_* \operatorname{div}_\Gamma \mathbf{v}$. Thus we have the expressions [3]

$$\lambda_\Gamma = 2\mu_* h, \quad \mu_\Gamma = \mu_* h, \quad \varkappa_\Gamma = \varkappa_* h. \tag{3.1}$$

We note that consideration of λ_Γ, μ_Γ, and κ_Γ in the defining expressions (2.5) is necessary in those cases where $\mu_* \gg \mu$, $\kappa_* \gg \kappa$ (for example for amorphous materials of the glass type the viscosity μ_* increases greatly with reduction in θ).

We will demonstrate that Eq. (2.6) at h = 0 yields conditions on the interphase boundary. In fact, if $\tau(h, \theta) \to \sigma(\theta)$ as $h \to 0$, it immediately follows from Eq. (2.3) that (with the prime denoting differentiation), $\varepsilon(h, \theta) \to \sigma(\theta) - \theta\sigma'(\theta)$, $\alpha(h, \theta) \to -\theta\sigma''(\theta)$. Since μ_* and κ_* in a thin phase mixture are of the order of magnitude of the viscosity and thermal conductivity of the phases themselves, then in the limit λ_Γ, μ_Γ, and κ_Γ vanish according to Eq. (3.1), leading to the conditions

$$\begin{gathered} [(p - \lambda \operatorname{div} \mathbf{v})\mathbf{n} - 2\mu D \cdot \mathbf{n}] = \operatorname{div}_\Gamma \{\sigma(\theta) G_\Gamma\}, \\ -\theta\sigma''(\theta)\dot{\theta} = \theta\sigma'(\theta) \operatorname{div}_\Gamma \mathbf{v} + [\varkappa \partial\theta/\partial n], \end{gathered}$$

which define the thermocapillary convection model [2]. In the present study the energy condition was derived for the surface Γ at rest, which permits calculation of $\dot{\theta}$ in the form $\partial\theta/\partial t + \mathbf{v} \cdot \nabla_\Gamma\theta$. On a moving surface Γ we have $\dot{\theta} = \delta\theta/\delta t + \mathbf{v} \cdot \nabla_\Gamma\theta$, where $\delta\theta/\delta t = \partial\tilde{\theta}/\partial t + \mathbf{v} \cdot \mathbf{n}\partial\tilde{\theta}/\partial n$, $\tilde{\theta}$ being the arbitrary smooth extension of θ in the vicinity of Γ. In fact, the derivative $\delta\theta/\delta t$ is equal to the derivative of θ along the vector field $(\mathbf{v}\cdot\mathbf{n}\mathbf{n}, 1)$, tangent to the trajectory of the surface Γ in the space $(\mathbf{x}, t)$, and is independent of the extension of $\tilde{\theta}$.

We note that in view of the equality $\operatorname{div}_\Gamma(\sigma G_\Gamma) = \nabla_\Gamma\sigma - \sigma k\mathbf{n}$ from the dynamic condition there follows the Laplace expression for the pressure change. Thus, $\sigma(\theta)$ is the interphase boundary surface tension coefficient and coincides with the free energy. Correspondingly, the entropy per unit area of Γ is equal to $-\sigma'(\theta)$, so that for a pure phase boundary the inequality $\sigma'(\theta) < 0$ is valid.

4. We will consider the problem of small perturbations of a spherical liquid film within which a polytropic gas is contained. More precisely, we will seek a solution of system (2.6), taking $\mathbf{g} = 0$, $\mathbf{n}\cdot[T] = p\mathbf{n}$, $\mathbf{n}\cdot[\mathbf{q}] = 0$, where $p = p_0(V_0/V)^\gamma$ is the average pressure, $V = \frac{1}{3}\int\limits_\Gamma \mathbf{x}\cdot\mathbf{n}d^2x$ is the gas volume, γ is the polytropy index, and the normal $\mathbf{n}$ is directed outward. In this case the first two equations of Eq. (2.6) lead to conservation laws

$$\frac{d}{dt}\int\limits_\Gamma h d^2x = 0,\quad \frac{d^2}{dt^2}\int\limits_\Gamma h\mathbf{x}d^2x = 0. \tag{4.1}$$

By choosing the proper Galilean coordinate system we ensure the equality

$$\int\limits_\Gamma h\mathbf{x}d^2x = 0 \tag{4.2}$$

and assume that Γ is stellate relative to zero. Then we can perform a stereographic projection $\mathbf{x} = r(\mathbf{s}, t)\mathbf{s}$ ($\mathbf{s}$ is a point of the unit sphere S) and in view of the first condition of Eq. (1.3),

$$\mathbf{v} = \dot{\mathbf{x}} = r\dot{\mathbf{s}} + (r_t + \dot{\mathbf{s}}\cdot\nabla_S r)\mathbf{s} = \mathbf{u} + (r_t + r^{-1}\mathbf{u}\cdot\nabla_S r)\mathbf{s}.$$

Here $\mathbf{u} = r\dot{\mathbf{s}}$ is the velocity field component tangent to S, by means of which in the function $f(\mathbf{s}, t)$ we calculate the total time derivative $\dot{f} = f_t + r^{-1}\mathbf{u}\cdot\nabla_S f$; the subscript corresponds to the partial derivative with respect to time. Thus, system (2.6) is reduced to the problem of finding the functions $r(\mathbf{s}, t)$, $\mathbf{u}(\mathbf{s}, t)$, $h(\mathbf{s}, t)$, and $\theta(\mathbf{s}, t)$. We note that, in view of the zero value of the viscosity and thermal conductivity coefficients of an ideal gas the last conditions of Eq. (1.3) cannot be insured without distinguishing boundary layers.

The spherically symmetric solution $r(t)$, $\mathbf{u} = 0$, $h(t)$, $\theta(t)$ in view of the equalities $\mathbf{v} = r_t\mathbf{s}$, $T_\Gamma = (\tau + 6\mu_* h r_t/r)G_S$ satisfies the ordinary differential equations $(hr^2)_t = 0$, $\rho_* h r_{tt} = p - 2(\tau + 6\mu_* h r_t/r)/r$, $\alpha\theta_t = 2\theta(\partial\tau/\partial\theta)r_t/r + 12\mu_* h(r_t/r)^2$ and initial conditions $r = r_0$, $r_t = r_1$, $h = h_0$, $\theta = \theta_0$ at $t = 0$. Here the functions $\tau(h, \theta)$, $\alpha(h, \theta)$ are specified and $p = p_0(r_0/r)^{3\gamma}$. It is obvious that a steady-state solution is possible for $p_0 = 2\tau_0/r_0$, $r_1 = 0$ (the zero subscript denotes characteristics of the equilibrium state). We will investigate its stability relative to initial perturbations of r, h, and θ.

We choose for length, time, velocity, and temperature scales the quantities r_0, $\mu_* r_0/\tau_0$, τ_0/μ_*, and θ_0, respectively, and introduce the dimensionless numbers

$$\delta = \frac{h_0}{r_0},\quad W = \frac{\rho_* r_0\tau_0}{\mu_*^2},\quad M = \frac{\alpha_0 r_0\tau_0}{h_0\mu_*\varkappa_*},$$

$$L = \frac{\tau_0}{\theta_0\alpha_0},\quad K_h = -\frac{r_0}{\tau_0}\left(\frac{\partial\tau}{\partial h}\right)_0,\quad K_\theta = -\frac{r_0\theta_0}{h_0\tau_0}\left(\frac{\partial\tau}{\partial\theta}\right)_0.$$

We take $r = 1 + R$, $\mathbf{u} = \nabla_S U$, $h = \delta(1 + H)$, $\theta = 1 + \Theta$, and linearize the system (2.6), (4.1), (4.2) for the basic solution, assuming the functions $R(\mathbf{s}, t)$, $U(\mathbf{s}, t)$, $H(\mathbf{s}, t)$, and $\Theta(\mathbf{s}, t)$ to be small and of the same order (the linearized equations admit potential perturbations of the velocity field). As a result, we obtain the problem

$$\begin{gathered}
H_t + 2R_t + \Delta_S U = 0,\\
WU_t = \Delta_S U + 2U - (3H_t + K_h H + K_\theta\Theta),\\
\delta W R_{tt} = \Delta_S R + 2R + 2\delta(3H_t + K_h H + K_\theta\Theta) - 6\gamma R_0,\\
\Theta_t = \delta L K_\theta H_t + M^{-1}\Delta_S\Theta,
\end{gathered}$$

$$\int\limits_S (H+2R)\,d^2s = 0,\quad \int\limits_S (H+3R)\,s\,d^2s = 0,\quad R_0 = \frac{1}{4\pi}\int\limits_S R\,d^2s,$$

in which separation of variables is possible. Thus, let $R(s, t) = R_m(s)\exp(\omega t)$, etc. [$R_m(s)$ is a spherical function of degree m], then the dispersion relationship takes on the form $F_m(\omega) = 0$. Here,

$$\begin{gathered}F_m(\omega) = [\omega + M^{-1}m(m+1)]\{(\delta W\omega^2 + m^2 + m - 2)\times\\ \times[W\omega^2 + 2(2m^2+2m-1)\omega + K_h m(m+1)] +\\ + 4\delta\omega(3\omega + K_h)(W\omega + m^2 + m - 2)\} +\\ + \delta L K_\theta^2\omega\{\delta W(m^2+m+4)\omega^2 + (m^2+m-2)[4\delta\omega + m(m+1)]\},\quad m \geqslant 2,\\ F_1(\omega) = (\omega + 2M^{-1})[W\omega^2 + 6(3\omega + K_h)] + 6\delta L K_\theta^2\omega,\\ F_0(\omega) = \delta W\omega^2 + 12\delta\omega + 2(3\gamma - 1) + 4\delta(K_h + \delta L K_\theta^2).\end{gathered}$$

Let $K_h = K\delta$ and $\delta \to 0$, whereupon it is valid to expand the root ω_m with largest real component in powers of a small parameter:

$$\omega_m = -\frac{m(m+1)K\delta}{2(2m^2+2m-1)}\left\{1 + \frac{M\delta}{4(2m^2+2m-1)}\left[\left(1 + \frac{m(m+1)W}{M(2m^2+2m-1)}\right)K - LK_\theta^2\right]\right\} + O(\delta^3),\quad m \geqslant 2,$$

$$\omega_1 = -\frac{K\delta}{3}\left\{1 + \frac{M\delta}{12}\left[\left(1 + \frac{2W}{9M}\right)K - LK_\theta^2\right]\right\} + O(\delta^3),$$

$$\mathrm{Re}\,\omega_0 = -6/W \text{ for } \gamma > 1/3.$$

From these considerations there follows the expected asymptotic stability of a homogeneous spherical film in the linear approximation for positive K (thermodynamic stabilization mechanism). Also evident is the destabilizing role of the dependence of τ on θ, described by the parameter LK_θ^2.

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