$$\int d\mathbf{k}_{1\ldots6} J \int_{-\infty}^{t_0} d\tau R_4(\tau) \exp i\left[(t-\tau)\sum_{n=1}^{4}\omega_n\right]\Phi_s \qquad (9)$$

where Φ is the sum of bilinear combinations of matrix elements and the quadruple integral in the wave field momenta estimated for $t/\tau_0 \to \infty$ yields the asymptotic $(\tau_0/t)^8$ for (9). The characteristic constant τ_0 is the greatest value of the integrand in (9) at the boundary points $\mathbf{k}_{1,2}$ of the inertial interval of the wave numbers $\mathbf{k}_1 \leq \mathbf{k} \leq \mathbf{k}_2$. In the general case, the last component in the right side of (7) describes interaction of four surface waves with the Fourier component of the field $\mathbf{v}$, most effective for mutual resonance. This component, additional to the collision integral [1, 2], models the nonlinear mechanism of the nonlocal response, in time, of the system of surface waves to the nonstationary inhomogeneous perturbation. Let us note that if the scale of the homogeneity of the flow considerably exceeds the wavelength then its influence can be taken into account by passage to a moving coordinate system [6]. In such an approximation the correction to the δ correlativity $M_{\mathbf{kk}'}$, which can be substantial for $\mathbf{k} \simeq \mathbf{k}'$, is not taken into account successfully.

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DEFORMATION AND BREAKUP OF A LIQUID FILM UNDER THE ACTION OF THERMOCAPILLARY CONVECTION

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Thermocapillary convection which develops in thin nonisothermal liquid layers produces significant deformation of the free surface [1-5]. This problem was considered in [1] within the framework of a model which neglected capillary pressure. A solution was obtained in [2] for the special case of harmonic temperature distribution; the problem was solved in an approximation linear in temperature perturbation. A more general formulation was considered in [3], where the equation of the free surface was found in an approximation analogous to the boundary layer approximation. The present study will offer new experimental results and define conditions under which thermocapillary convection in a liquid leads to breakup of the film into individual drops.

1. If the plane upon which a thin film of liquid is deposited is oriented perpendicular to the acceleration of gravity then in dimensionless variables the equation of the free surface will have the form [3]

$$\xi^2 + \xi'^2 - 2\xi\xi'' + \varepsilon\vartheta(x) = C, \qquad (1.1)$$

where $\xi(x)$ is the local thickness of the liquid layer; $\vartheta(x)$, the temperature of the free surface; C, a constant defined from additional conditions; $\varepsilon = 3\Delta T\sigma_T'/\sigma_0$; σ_0, the mean surface tension coefficient: $\sigma_T' = |d\sigma/dT|$; ΔT, the characteristic temperature difference, for example, between the hotter and colder parts of the layer, per

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TABLE 1

Liquid	$\rho \cdot 10^{-3}$, kg/m^3	$\eta \cdot 10^3$, (kg/m·sec)	$\sigma_0 \cdot 10^3$, N/m	$\sigma_T' \cdot 10^3$, N/(m·K)	$p \cdot 10^{-2}$, Pa
Ethyl alcohol	0,8078	1,200	22,80	0,083	57,14
n-heptane	0,6836	0,414	20,86	0,101	47,24
n-decane	0,7299	0,907	23,89	0,092	1,200
n-undecane	0,7394	1,182	24,78	0,089	0,345

unit length. For the unit of length we take the capillary radius $(\sigma_0/\rho g)^{1/2}$ (where ρ is density and g is the acceleration of gravity).

Under reduced gravitation conditions the term ξ^2 in Eq. (1.1) can be neglected if the layer length L is small in comparison to the capillary radius, since $(\xi/\xi')^2 \sim L\rho g/\sigma_0 \ll 1$. Under terrestrial conditions the capillary radius does not exceed 1-3 mm and the surface relief is essentially determined by the weight force (first term on the left of Eq. (1.1)). However at the edges of the film and in the state preceding breakup ($\xi \to 0$) the remaining terms prove significant and cannot in general be neglected.

To obtain the conditions for film breakup we will consider a model problem with harmonic temperature distribution along the x-coordinate: $\vartheta(x) = 0.5 \cos kx + \text{const}$. As comparison with experiment and results obtained with other temperature distributions proves, the concrete form of the function $\vartheta(x)$ is of little significance. We will consider the length of the layer to be sufficiently great that boundary effects can be neglected. The dimensionless thickness of an isothermal film is equal to H. For sufficiently small values of the parameter $\varepsilon/8H^2$ the solution of Eq. (1.1) is the function

$$\frac{\xi}{H} = 1 - \frac{\varepsilon \cos kx}{4H^2(1+k^2)} - \frac{\varepsilon^2 \cos 2kx}{64H^4(1+k^2)(1+4k^2)} - \frac{\varepsilon^3}{512H^6(1+k^2)(1+4k^2)}\left[\frac{1+7k^2}{1+k^2}\cos kx + \frac{1+3k^2}{1+9k^2}\cos 3kx\right] - \ldots \quad (1.2)$$

This solution allows determination of the approximate condition for film breakup. Film breakup sets in if the parameter ε exceeds some critical value ε_*. In the limit $k \ll 1$ (spatial period of temperature modulation large as compared to capillary radius) we have

$$\varepsilon_* \approx 3H^2 \quad (1.3)$$

or in dimensional form $\Delta T_* \sigma_T' \approx \rho g h^2$ (h is the dimensional mean thickness of the liquid layer).

In the other limit ($k \gg 1$, low accelerations, "shortwave" modulation), breakup of the liquid layer sets in at $\varepsilon_* \approx 3.6H^2k^2$ or $\Delta T_* \sigma_T'/\sigma_0 \approx 4.8(\pi h/\lambda)^2$ (where λ is the spatial period of the temperature). Thus, if $\lambda \geqslant 10$ cm, breakup of an ethyl alcohol layer with mean thickness 0.5 mm should set in even at $\Delta T_* \sim 0.3$ K, i.e., for very slightly nonisothermal conditions. A layer of the same medium 5 mm thick breaks into individual droplets at $\Delta T_* \sim 30$ K, etc. It should be understood that these estimates are valid only while the liquid layer can be considered thin and the flow slow.

It follows from Eq. (1.2) that in the approximation linear in ε the maximum height differential $\Delta\xi$ between hot and cold portions is inversely proportional to the initial layer thickness H:

$$\Delta\xi = \varepsilon/2[H(1+k^2)], \quad (1.4)$$

which coincides with the conclusions of [2].

2. To study breakup conditions experimentally, a thin liquid layer with free upper surface was deposited on a horizontal rectangular metal plate measuring 70 × 74 × 3 mm. The sharp edges of the plate hindered liquid spreading. The two opposite ends of the plate were cooled, while the midsection (along x) of the plate was heated by heat exchangers connected to a high stability thermostat. As a result the temperature in the liquid layer decreased with x from the midsection to the ends in a nearby linear manner. Temperature measurements were performed with copper–Constantin thermocouples fitted into the plate to an accuracy of 0.1 K.

Surface relief was studied with an optical cathetometer as well as a short focus shadow apparatus equipped for operation with reflected light. Measured values of the inclination of the surface were integrated graphically to define its form. The working liquids used were ethyl alcohol, heptane, decane, and undecane. Values of density ρ, viscosity η, surface tension σ_0, the temperature coefficient of surface tension σ_T', and saturated vapor pressure at 20°C were taken from [6, 7] and are shown in Table 1. Liquid layer thickness was varied from 0.1 to 1.5 mm. Temperature changes between heater and cooler did not exceed 30 K.

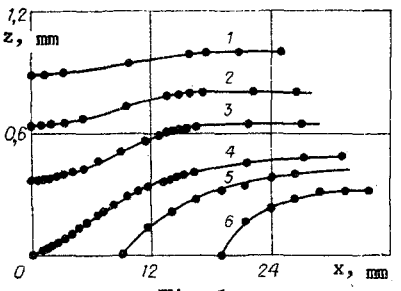

Fig. 1

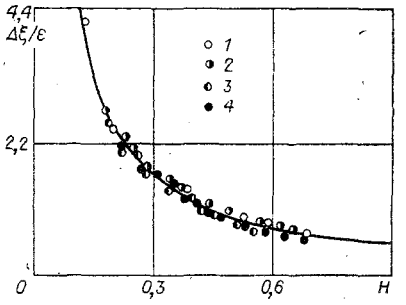

Fig. 2

Figure 1 shows the experimentally measured local thickness z of a layer of n-decane as a function of horizontal coordinate measured from the heater. The temperature head was 11.5 K. The presence of the temperature head stimulates a steady-state thermocapillary convection (gravitational convection in such thin films is insignificant [8]). Liquid motion proves to be symmetric about the middle part of the layer and is directed from the hot to the cold areas on the surface. As a result of this flow the initially plane liquid layer deforms, its thickness above the heat source becoming smaller than at the coolers. The curves of Fig. 1 correspond to various amounts of liquid poured onto the plate (initial thickness of the undeformed layer h for curves 1-6 equal to 0.95, 0.74, 0.53, 0.32, 0.29, and 0.14 mm). It is evident that for small layer thickness (curves 4-6) breakup of the liquid layer and exposure of the bare heated segment of the bottom occur. The liquid collects as a thin drop above the cooled portion of the substrate. We will note that a nonzero dynamic boundary angle then exists. The cause of this dynamic angle is thermocapillary motion. Temperature equalization leads to a rapid spreading of liquid over the entire plate.

The maximum height differential Δz between hot and cold portions of the layer proves to be proportional to the temperature differential ΔT between the heater and cooler and inversely proportional to the initial layer thickness h. The experimental results are shown in dimensionless form in Fig. 2. The liquid used was ethyl alcohol. Experimental points 1-4 were obtained at $\Delta T = 5.6$; 12.5; 14.2; 18.4 K, and the line was constructed with Eq. (1.4). As is evident from the figure, this equation describes the deformation of thin liquid layers well over the entire range of parameters H, ε studied. Points obtained under various temperature conditions lie on one universal curve.

Critical values of the parameter ε at which breakup of a liquid film of initial thickness H occurs are shown in Fig. 3, where the line corresponds to Eq. (1.3). The same graph shows experimental points (1-3), obtained in experiments with heptane, decane, and ethyl alcohol, respectively. The experimental results agree well with theory, despite the approximate character of the solution. We note that the liquids used (see Table 1) differ in dynamic viscosities by a factor of three. However, as is evident from the graph, and also from Eqs. (1.1), (1.3), liquid viscosity does not affect the amount of deformation or the liquid layer breakup condition.

3. The analytical problem for a film with two free surfaces located under conditions of weightlessness ($k \gg 1$) reduces to solution of the equation

$$3\xi'^2 - 6\xi\xi'' + 2\varepsilon\theta(x) = C. \tag{3.1}$$

We locate the origin of the coordinate system in the center of the undeformed layer. Equation (3.1) differs from Eq. (1.1) in numerical coefficients and the absence of a term $\sim\xi^2$. The changes in the coefficients are due to changes in boundary conditions and the velocity profile. Film breakup occurs for the condition $\Delta T \geq \Delta T_*$:

$$\Delta T_* \approx \frac{3}{2}(\sigma_0/\sigma_T')\left(\pi\frac{h}{\lambda}\right)^2. \tag{3.2}$$

In the experiments a liquid layer with two free surfaces was formed by a thin liquid film supported on a wire frame in the horizontal plane. The width of the frame d = 3 mm, which is comparable to the value of the capillary radius; therefore such a film proves to be stable and can exist over the course of several hours. The length of the frames L = 24 mm. The opposite ends of the frame were held in thermal contact with a heater and cooler, permitting creation of a temperature differential along the film. Type TÉMO-6 semiconductor microcoolers were used for the cooler and heater. Temperature measurements were performed with thermocouples with junctions introduced directly into the liquid layer. The liquid used was n-undecane, which is distinguished by its low vapor pressure (see Table 1), which permits neglect of liquid evaporation.

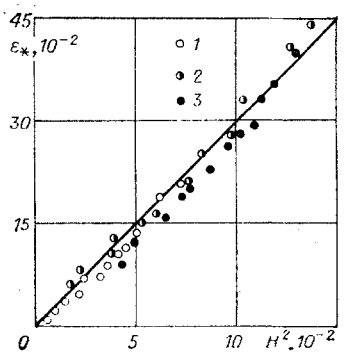

Fig. 3

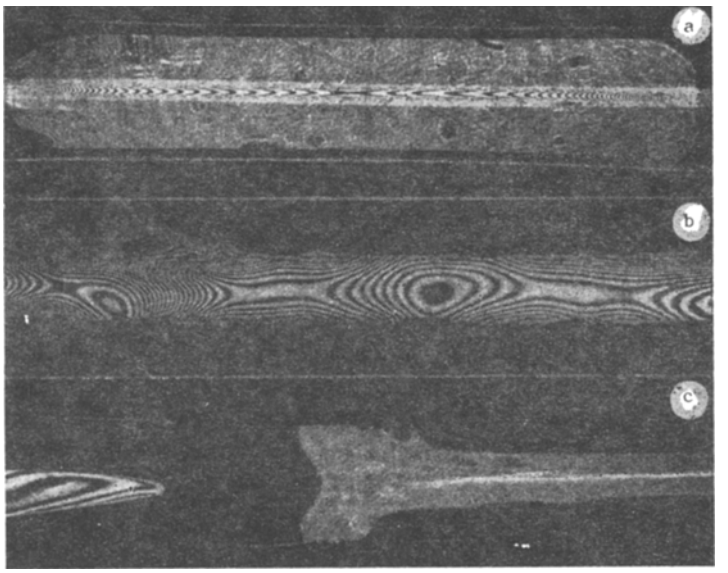

Fig. 4

The liquid film was illuminated from above by a parallel beam of monochromatic (laser) light. As a result a microscope viewing light reflected from the film showed an interference pattern – bands of equal thickness, created by rays reflected from the upper and lower surfaces of the liquid. The interference patterns obtained are shown in Fig. 4.

If the liquid film is not planar, but has a concave or convex form with respect to width, the interference bands are visible only in a narrow portion of the surface and indicate inhomogeneity of the film over width (Fig. 4a). Then by increasing or decreasing the quantity of liquid a situation can be achieved in which the upper and lower free surfaces of the film have minimum curvature and are parallel to each other. In this case wide interference bands are observed over practically the entire surface (Fig. 4b). Since the transition from one interference band to the next corresponds to a change in film thicknesses by 0.2 μm, thickness inhomogeneity over the entire film area did not exceed 0.01 mm. It is evident that in this case the liquid film thickness can, with good accuracy, be considered equal to the thickness of the wire frame. In the experiments wires with diameters from 0.15 to 0.7 mm were used, thus permitting variation of the film thickness over the same limits.

After switchon of the cooler and heater a thermocapillary flow commences and the film thickness near the cooler becomes greater than near the heater. As a result of film deformation the upper and lower surfaces become significantly nonparallel and the interference pattern disappears. With further heating of the liquid layer the deformation intensifies, the liquid film thickness near the heater grows smaller, and upon attainment of some critical temperature difference ΔT_* the film breaks up. The interference pattern appears again only directly before film breakup at the hottest point, when the film thickness comprises only a few wavelengths of the laser light (Fig. 4c).

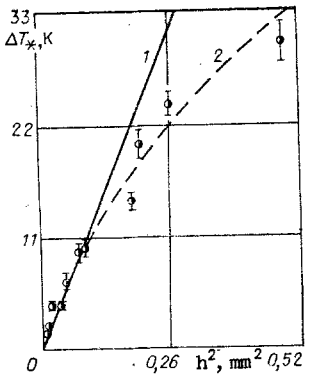

Fig. 5

The critical temperature differences between the cold and hot ends of the film which cause its breakup are shown as functions of the square of initial thickness in Fig. 5. According to Eq. (3.2) destruction of a two-sided liquid film of thickness, for example, 0.5 mm, should occur at $\Delta T_* \sim 0.5$ K. As is evident from the graph, in experiment the film proves to be stable at significantly higher temperature differences. This divergence can be explained by the fact that the real film is not infinite in width, as was assumed in the analytic solution, but rather is bounded by the wire frame. As a result the major portion of the capillary pressure in the liquid is created by the curvature of the surface across the liquid layer. We will attempt to consider this effect.

For a change in film thickness by an amount Δh the capillary pressure within the liquid changes by $\Delta p = 4\sigma_0/(h^2 + d^2)$ and in Eq. (3.2) an additional term appears:

$$\Delta T_* \approx \frac{h^2\sigma_0}{\sigma_T'}\left(\frac{3}{2}\pi^2\frac{1}{\lambda^2} + \frac{4}{h^2+d^2}\right). \tag{3.3}$$

In Fig. 5, Eq. (3.3) is shown by the line 1, and the experimental points by line 2. It is evident that the experimental points agree with Eq. (3.3) for small layer thicknesses ($h \leq 0.3$ mm), for which the ratio of film thickness to width is ~ 0.1. For thicker ($h \geq 0.3$ mm) layers, the film width is comparable to its thickness and the simple considerations presented above prove inapplicable. In this case the form of the lower free surface will be affected more intensely by gravity and Rayleigh–Taylor instability will have a greater role in film breakup. As a result collapse of the liquid layer occurs significantly earlier than for pure thermocapillary instability.

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