Self-oscillations of a liquid near a free surface during continuous local heating

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(Submitted 30 January 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **49**, No. 6, 330–333 (25 March 1989)

An instability of Marangoni convection during the local heating of a liquid by a laser beam has been studied experimentally. A theoretical explanation is offered.

The processes by which flows in liquids become unstable during heating by laser beams have recently attracted considerable interest. In particular, in Refs. 1 and 2 an absorbing liquid was placed in a flat vessel, and the liquid was heated from below by the focused beam from an argon laser. The threshold laser power for excitation of the oscillations in the liquid was ≈ 1 W. A model of a dissipative structure (a "circulator") was proposed in Ref. 2 in an effort to explain the oscillations in the liquid flow.

In the present letter we report an experimental study of, and offer an explanation for the self-oscillations which occur in the flow of a liquid when laser light is focused near the free surface of the liquid (Fig. 1a), with an excitation threshold of a few milliwatts. To study the self-oscillations, we used ethyl alcohol colored with iodine (with an absorption $\sim 5–10\ \mathrm{cm}^{-1}$). The structure of the liquid flows was observed in a rectangular cell 1 mm thick and 4 mm wide, in the arrangement shown in Fig. 1. The flows were visualized by adding particles of aluminum powder to the liquid. After the laser beam was turned on, at a power level below the instability threshold, the liquid remained immobile for ~ 0.5 s; then a steady-state flow was established abruptly. The structure of this flow is shown in Fig. 1b. The liquid tended to leave the heating zone in the direction toward the free surface at a velocity of a few centimeters per second and then spread outward along the surface in various directions.

The instability of the liquid motion was manifested in the following way: In a certain interval of the laser beam power, and in a certain interval of the distance from

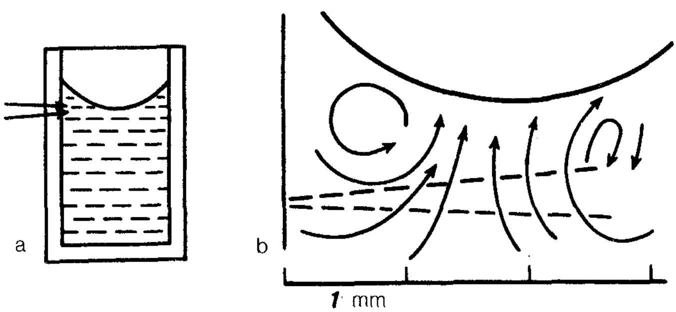

FIG. 1. a—Arrangement for fucusing the cw beam from an argon laser (wavelength of 0.488 μm) into the end of a rectangular cell; b—pattern of the motion in the liquid observed through a microscope (the dashed lines show the light beam).

the heating zone to the surface, the motion underwent a transition from a steady flow to an ordered periodic regime. The onset of oscillations in the flow velocity could be seen quite well both through direct observation of the structure of the flow and from the periodic changes in the structure of the transmitted beam, which was distorted by thermal defocusing. The oscillations were definitely nonlinear and consisted of a periodic ($\tau \approx 1$ s) sharp acceleration and deceleration of the liquid flow, in such a manner that the liquid remained essentially immobile over most of the period. The pattern of variations in the transmitted beam was of the nature of "heart beats."[3]

In the theoretical model of Ref. 2, a convective transport of heated liquid toward the surface plays an important role. In an effort to identify the role by convective transport in the present case, we placed the liquid in a capillary with an internal diameter of 2 mm. Both the characteristics of the oscillations and the structure of the liquid flow remained essentially the same when we positioned the capillary horizontally and even when we turned it over. In all cases we could clearly see a flow of liquid away from the heating zone toward the free surface. This circumstance is evidence that convection in the liquid is of minor importance in our particular experimental arrangement.

To interpret the experiment, we consider a model of a dissipative structure (a "thermillator") which describes the heat and mass transfer in a liquid in a current tube l, which has a section near the free surface of the liquid, $l_1 < l < l_2$. The heating is assumed to be lumped at the point $l = l_0$. In this case it becomes necessary to consider heat conduction along the current tube. The equation describing the dynamics of the temperature (T) distribution along the current tube is, in this case ($0 \leqslant l \leqslant L$),

$$\frac{\partial T(l,t)}{\partial t} = \chi \frac{\partial^2 T(l,t)}{\partial l^2} - v(t)\frac{\partial T(l,t)}{\partial l} - \alpha [T(l,t) - T_0] + \kappa P(l_0), \quad (1)$$

where χ is the thermal conductivity, v is the flow velocity, α and κ are the heat-transfer coeffiecient and the heat evolution, and P is the power of the light which is incident on the system. The boundary conditions on the current tube are $T(0,t) = T_0$, $T(L,t) = T_0$.

The observed structure of the liquid flow is evidence that the processes which occur in the system are driven by thermocapillary forces at the free surface (Marangoni convection).[4] The effective force may change sign as liquid, which has not yet been heated in the zone acted upon by the laser beam, is carried to the surface; as a result, the flow may effectively accelerate and decelerate. The motion of the liquid in the current tube is described by the equation

$$\frac{\partial v(t)}{\partial t} = \xi [T(l_1,t) - T(l_2,t)] - \gamma v(t), \quad (2)$$

where ξ specifies the Marangoni force, and γ is the viscous friction coefficient. A numerical solution of Eqs. (1) and (2) demostrates that there is a region of self-oscillations. This region is bounded both from below and from above by critical values of $h = l_1 - l_0$ and P. The shape of the oscillations depends strongly on the distance from the heating zone to the free surface, h. At small values of h, for example, the

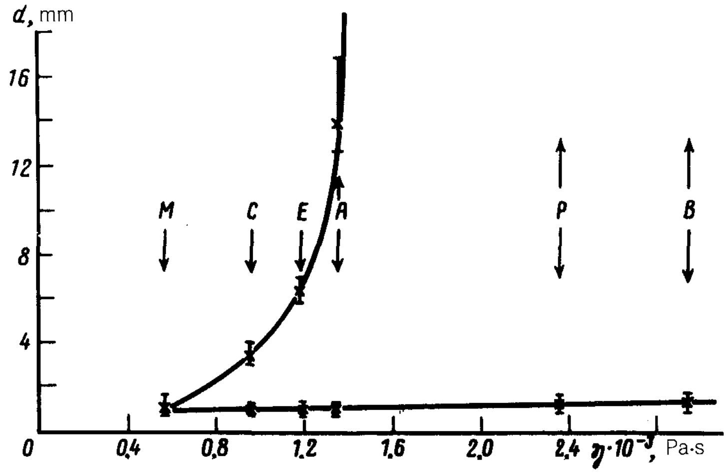

FIG. 2. Region in which the oscillatory instability of Marangoni convection exists in the plane of the parameters η (the dynamic viscosity of the liquid) and d (the cell diameter). *M*—Methyl alcohol; *C*—cyclohexane; *E*—ethyl alcohol; *A*—allyl alcohol; *P*—propyl alcohol; *B*—butyl alcohol. The values of η are taken at 20 °C.

oscillations are nearly sinusoidal; as h increases, they become relaxation oscillations, and their frequency decreases—in good qualitative agreement with the experimental results.

The role played the viscosity and the geometric dimensions of the cell was determined in a series of experiments in which we used a variety of solvents and cylindrical cells of various diameters. The region in which the oscillations exist is shown in Fig. 2. For methyl alcohol the oscillations died out after a few cycles; for cyclohexane, ethyl alcohol, allyl alcohol, undamped oscillations were observed in a limited interval of diameters. For solvents with a higher viscosity we were not able to establish an upper limit on the cell diameter.

Analysis of the effect of the parameters in this model also shows that the effect disappears as the thermal conductivity of the liquid increases. This conclusion is supported by the experiments: For water, whose viscosity falls in the region in which oscillations exist, there is absolutely no effect, because of the high thermal conductivity.

In summary, the model purposed here can describe the basic aspects of the behavior, and answers most of the questions posed in Ref. 5. In order to describe the effects which occur at high values of the adsorbed power, we would have to take account of the temperature dependence of the viscosity of the liquid. We might add that in several cases we observed a periodic creation of vortices near the main jet of the flow. The motion of these vortices modulated the oscillations of the jet velocity; as the heating power was raised, the oscillations become chaotic, and the flow turbulent. A study of the scenario for the onset of chaos in this system of interest for establishing a model for the processes which occur and also because this experiment is exceptionally simple and graphic.

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