Stable Localized Patterns in Thin Liquid Films

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(Received 4 November 1991)

We study a two-dimensional nonlinear evolution equation which describes the three-dimensional spatiotemporal behavior of the air-liquid interface of a thin liquid film lying on the underside of a cooled horizontal plate. We show that the equation has a Liapunov functional and exploit this fact to demonstrate that the Marangoni effect can stabilize the destabilizing effect of gravity (the Rayleigh-Taylor instability) allowing for the existence of stable localized axisymmetric solutions for a wide range of parameter values. Various properties of these structures are discussed.

PACS numbers: 68.15.+e, 47.20.Dr, 47.20.Ky, 68.90.+g

Ever since the discovery of solitons [1], there has been a great deal of interest in the existence of spatially localized solutions in nonlinear systems. In addition to the solitons of integrable systems, spatially localized solutions have been found in systems *with* dissipation such as the complex Ginzburg-Landau equation for systems near an inverted bifurcation [2–5] and a nonlinear phase equation [6,7]. On a somewhat different footing, we point out that nonlinear evolution equations which result from perturbation expansions have been found to be very useful in studying the dynamics of many physical systems. An example of such an equation is the well-known Kuramoto-Sivashinsky equation which describes systems such as liquid film flow [8–10] and flame front propagation [11]. Another such equation which results from similar derivations [12–15] is an equation which describes the fluid-air interface of a thin liquid film on the surface of a horizontal plate subjected to a vertical temperature gradient, which gives rise to the Marangoni effect. The Marangoni effect is found to be very important in low gravity conditions and therefore for processes of space technology [13]. The one-dimensional form of the equation has been found to exhibit localized solutions [14]. However, the existence of localized solutions for the two-dimensional equation is still an open question.

In this Letter we (1) show that the two-dimensional evolution equation, describing the three-dimensional spatiotemporal behavior of the air-liquid interface, has a Liapunov functional that is strictly nonincreasing in time; and (2) exploit this fact to demonstrate the existence of stable axisymmetric localized solutions for a wide range of parameter values. We note that similar techniques could be applied to other systems for which a Liapunov functional exists. We find that for given parameter values in the equation, the stable localized solutions exist for a wide range of amplitudes and that there is a minimum amplitude below which the solution is no longer stable, but damps to a flat state. Further we find that the amplitude of the localized solutions varies nearly linearly with the logarithm of the thickness of the film surrounding the localized state, larger amplitudes corresponding to a thinner film. This implies that the maximum amplitude of the localized state which can exist for given parameter values will depend on the noise in the system and on irregularities in the plate, since film rupture will be induced by lower noise levels and by smaller irregularities for thinner films. It will be very interesting to see whether experiments of a thin liquid film on the underside of a cooled horizontal plate will exhibit such stable stalactite-like structures.

The equation we study is

$$h_\tau+\nabla\cdot\left[\left(\frac{\gamma h^3}{3}-\frac{Bmh^2}{2(1+Bh)^2}\right)\nabla h\right]+\frac{s}{3}\nabla\cdot[h^3\nabla\nabla^2 h]=0\,. \tag{1}$$

Here $h(x,y,\tau)$ is the dimensionless thickness of the film; γ, s, and B are positive dimensionless parameters describing the gravity, surface tension, and the Biot number (related to the rate of heat transfer at the free surface), respectively, of the system; τ is a rescaled time; and $\nabla\equiv(\partial_x,\partial_y)$. Equation (1) results from making a thin-film approximation expansion of the incompressible fluid dynamics equations and assuming the surface tension is large.

This equation was derived in Refs. [12–15] and, assuming $\gamma>0$, $B>0$, and $s>0$, describes the spatiotemporal evolution of a thin liquid film bounded by a rigid horizontal plate of uniform temperature from above and by its free surface from below (the Rayleigh-Taylor instability); and experiencing surface tractions arising from the surface tension dependence on the temperature (the Marangoni or the thermocapillary effect). The value of the parameter m is 1 when the temperature of the plate is lower than that of the ambient air and -1 when the plate temperature is higher. A linear stability analysis shows that for $m=1$, a film of uniform thickness h_u is stable if $h_u(1+Bh_u)^2<3B/2\gamma$, demonstrating that the Marangoni effect can stabilize the Rayleigh-Taylor instability. For $m=-1$ the Marangoni effect is destabilizing.

Similar to the one-dimensional case [14], Eq. (1) may be written in the form

$$h_\tau=\nabla\cdot\left[h^3\nabla\left(\frac{\delta F}{\delta h}\right)\right], \tag{2}$$

where the Liapunov functional F is

$$F(\tau)=\int dx\,dy\left[P(h)+\frac{s}{6}||\nabla h||^2\right], \tag{3}$$

and the potential P is

$$P(h) = -\frac{\gamma}{6}h^2 + \frac{Bm}{2}h\ln\left(\frac{h}{1+Bh}\right). \tag{4}$$

In the above and below, $\|\ \|$ denotes the vector norm, and the domain of integration is assumed to be periodic. Therefore boundary terms will vanish.

Taking $dF/d\tau$ and integrating by parts gives

$$\frac{dF}{d\tau} = -\int dx\,dy\,h^3\left\|\nabla\left(-\frac{dP}{dh} + \frac{s}{3}\nabla^2 h\right)\right\|^2. \tag{5}$$

Since $h>0$, we have $dF/d\tau \le 0$ and thus F is strictly nonincreasing in time. We also note that $Q=\int dx\,dy\,h$ is a constant of the motion. Therefore, assuming that a stable stationary state exists, it will be given by minimizing F subject to the constraint Q is constant. Taking $\delta(F+\lambda Q)=0$, where λ is a Lagrange multiplier, we find

$$\frac{s}{3}\nabla^2 h = \frac{dP}{dh} + \lambda \tag{6}$$

for the equation satisfied by a stationary state. This equation may be solved by finding the steady-state solutions of

$$h_t = \frac{s}{3}\nabla^2 h - \frac{dP}{dh} - \lambda\,, \tag{7}$$

where the constraint Q is constant is imposed by taking

$$\lambda(t) = -\frac{1}{\int dx\,dy}\int dx\,dy\,dP/dh\,. \tag{8}$$

Defining $R \equiv dP/dh - (s/3)\nabla^2 h$ we have

$$dF/dt = -\int dx\,dy\,R^2 + \left[\int dx\,dy\,R\right]^2 \Big/ \int dx\,dy \le 0\,.$$

Therefore, the solutions of Eq. (6) found via solving the dynamical equation (7) are not only stationary solutions of Eq. (1), but are guaranteed to be *stable* stationary solutions since the Liapunov functional F is strictly nonincreasing during the evolution of Eq. (7) [16]. We stress that Eq. (7) does *not* reproduce the transient behavior of the system, but is only a mathematical trick to find stable stationary states. If one is interested in the time-dependent behavior, then it is necessary to solve Eq. (1), which requires a great deal more computer time and is more difficult to solve due to the highly nonlinear nature of the equation.

By taking $m=1$, and scaling with respect to time, space, and amplitude as $\tau=(8\gamma s/9B^3)T$, $(x,y)=\sqrt{s/\gamma}(X,Y)$, and $h=(3B/2\gamma)H$, Eq. (1) may be written as

$$H_T + \nabla\cdot\left[\left(H^3 - \frac{H^2}{(1+\beta H)^2}\right)\nabla H\right] + \nabla\cdot[H^3\nabla\nabla^2 H] = 0\,, \tag{9}$$

where $\nabla=(\partial_X,\partial_Y)$ and $\beta=3B^2/2\gamma$. To write Eqs. (2)-(8) in the scaled variables, make the substitutions $x\to X$, $y\to Y$, $h\to H$, $s\to 3$, $\gamma\to 3$, $Bm\to 2$, $B\to\beta$.

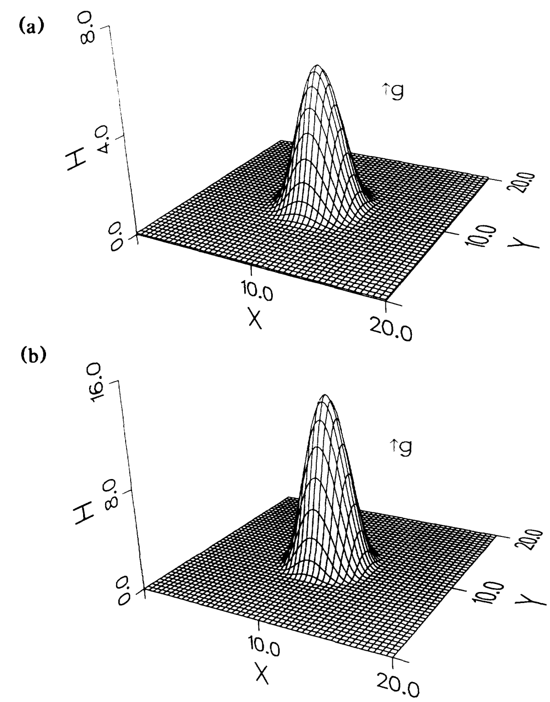

FIG. 1. Stable localized solutions of Eq. (9) for $\beta=1$. The arrow indicates the direction of gravity. Note that similar steady solutions of different amplitudes A are admitted by Eq. (9) for the same value of β. (a) $A=5.787$, $H_b=6.279\times10^{-2}$, $\lambda=1.951$; (b) $A=13.607$, $H_b=6.506\times10^{-3}$, $\lambda=4.054$.

We look for stable localized solutions of Eq. (9) by numerically solving Eqs. (7) and (8) in the scaled variables. We used fourth-order finite differencing for the spatial derivatives, taking a 92 by 92 grid, and used the Euler method for the time integration (since we are only interested in the stationary state, the Euler method is entirely sufficient). Periodic boundary conditions were used. The accuracy of the solution was checked by evaluating dF/dt from Eq. (5).

Figure 1(a) shows a stable localized solution for $\beta=1$ which evolved from an initial Gaussian. The amplitude of this state is $A=5.787$ and the background thickness of the film surrounding the state is $H_b=6.279\times10^{-2}$. From Eq. (8) we find $\lambda=1.951$. Figure 1(b) shows a stable localized solution with amplitude different from that of Fig. 1(a) for the same value of β (note the change in vertical scale). For this state we have $A=13.607$, $H_b=6.506\times10^{-3}$, and $\lambda=4.054$. Note that the larger amplitude state has a smaller background thickness. To demonstrate that stable localized solutions exist for a wide range of β and that the form of the solutions are very similar, Figs. 2(a) and 2(b) show stable localized states for $\beta=0.01$ and $\beta=100$, respectively (again note the vertical scale). For Fig. 2(a) we have $A=12.990$, $H_b=6.068\times10^{-2}$, and $\lambda=1.864$. For Fig. 2(b) we have $A=2.186$, $H_b=3.355\times10^{-3}$, and $\lambda=5.241$. We see that the width of the localized state is rather insensitive to the

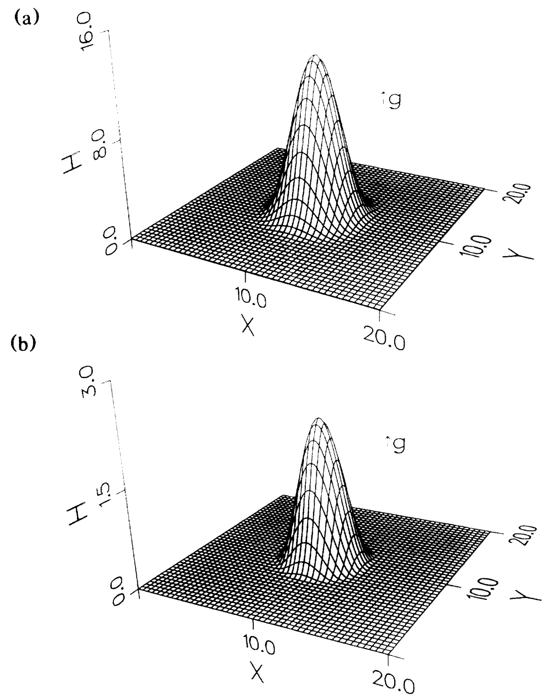

FIG. 2. Stable localized solutions of Eq. (9) for various values of β: (a) $\beta=0.01$, (b) $\beta=100$. Note the change in vertical scale.

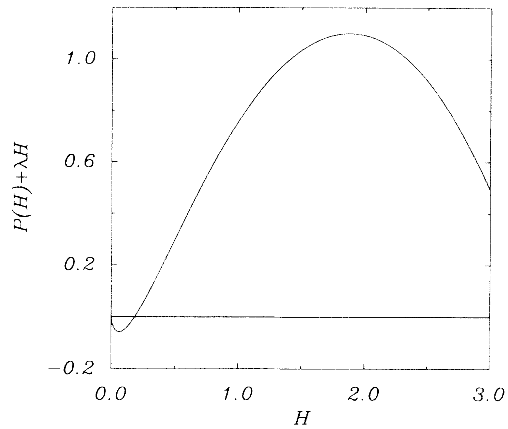

FIG. 3. The effective potential $P(H)+\lambda H$ for the localized state shown in Fig. 1(a), as a function of H. The value of $\lambda=1.951$ is calculated using Eq. (8). The background thickness $H_b=6.279\times 10^{-2}$ lies in the minimum of the potential well.

value of β.

Some insight may be gained by plotting the effective potential $P(H)+\lambda H$ as a function of H. Figure 3 shows such a plot for the state shown in Fig. 1(a). We see that there is a potential well (or basin of attraction) which confines the flat portion of the film surrounding the localized state. If the flat portion of the solution is perturbed (with a perturbation of zero mean), it relaxes back to the minimum of the potential. As H increases, the potential increases and reaches a maximum and then decreases, approaching $-\infty$ as $H\to\infty$. In contrast to the localized states in phase dynamics [6,7] where *two* potential wells exist, there is only *one* potential well here. This implies that stable kink solutions are not possible [7]. Another way of interpreting the potential for the 1D equation is in interpreting the spatial coordinate as a time. Then the 1D version of Eq. (6) is simply Newton's law with $s/3$ equal to the mass and $dP/dh+\lambda$ equal to a force. The potential energy corresponds to $V=-P-\lambda h$, the total energy being a constant of the motion. A 1D localized solution corresponds to a particle starting near the local maximum of V, sliding down the hill, up the other side, and then returning. To interpret the 2D axisymmetric solution in this fashion it is necessary to write $\nabla^2 h$ as $(1/r)(rh_r)_r$ (using polar coordinates where $r=0$ corresponds to the maximum of the solution). Then Newton's law is $(s/3)(h_{rr}+h_r/r)=-V_h$, where r is interpreted as the time and $1/r$ is a time-dependent frictional coefficient. The axisymmetric solution corresponds to a particle starting at a value of h and r near the maximum of the solution, sliding down the hill, and up the other side approaching the local maximum of V and staying there. Note that, although these analogies with a particle in a potential are useful, they say nothing about the *stability* of the solutions.

Figure 4 shows plots of A as a function of $\log_{10}(H_b)$ for stable localized solutions for five different values of β. To produce these plots it was first necessary to determine the minimum amplitude (for each value of β) for which stable localized solutions exist. This was done by decreasing the amplitude of a stable state by a small amount and allowing the system to come to equilibrium. This process was repeated until at some amplitude the solution was no longer stable, but damped to a flat state. Performing this process for each value of β then determines the end points at the lower right of the curves. To the lower right of these end points no stable localized solutions exist. The next step is to gradually increase the amplitude, beginning at the right-hand end points and allowing the system to come to equilibrium at each step. As H_b became very small, it was necessary to decrease the time step in order to get an accurate solution (determined by evaluating dF/dt). Therefore, the upper left end points of the curves only reflect a finite amount of computer time and not the nonexistence of solutions. We expect that the curves continue to the upper left at approximately constant slope.

An important point that can be inferred from Fig. 4 is that even though stable localized solutions may exist for the deterministic Eq. (1) for all amplitudes greater than some minimum, in practice—at some sufficiently large amplitude—the film surrounding the localized state will become sufficiently small so that rupture will be induced by noise in the system or irregularities in the plate. This implies that there is a maximum amplitude, the value of which will be determined by the noise level or by the

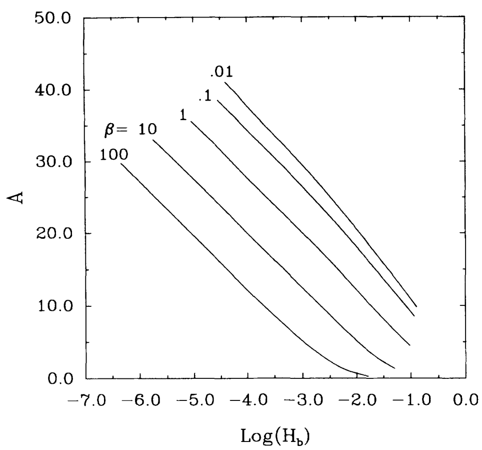

FIG. 4. The amplitude A of the stable localized state as a function of the logarithm of the background thickness, $\log_{10}(H_b)$, for various values of β. Note that these curves are nearly linear.

magnitude of the irregularities. More precisely, we would expect that the maximum amplitude would vary approximately linearly with the logarithm of the magnitude of the noise or irregularities. Even if the plate is perfectly smooth and all external vibrations are eliminated, *noise-induced rupture* would still result from thermal noise, so that in any physical system there will always be a maximum amplitude for which stable localized solutions can exist.

Although the two-dimensional equation is our primary concern in this paper and since there was very little discussion about the localized solutions in Ref. [14], we note a few properties of the one-dimensional solutions. First, just as for the 2D equation, we find that stable localized solutions exist for a wide range of parameter values. One difference between the 1D and 2D solutions is that for a given background thickness, the amplitude of the 1D solution is smaller than that of the 2D solution, which is consistent with interpreting the 2D and 1D solutions as the motion of a particle in a potential well with and without friction, respectively. For example, for the potential of Fig. 3 the amplitude of the 1D solution is 3.422 as compared to 5.787 for the 2D solution [Fig. 1(a)]. Another difference is that for a given value of β the maximum thickness of the background state for which the solution is still stable and does not damp to a flat state is larger for the 1D solution than for the 2D solution. For example, for $\beta=1$ the critical H_b for the 1D and 2D solutions are about 0.24 and 0.10, respectively. Another interesting property is that, just as for the 2D solutions, the amplitude A varies nearly linearly with the logarithm of the background thickness H_b, although the slope is different. In fact for the 1D solution it is straightforward to show from the potential that in the limit of large A and small H_b, $A=-2[1+\ln(\beta H_b)]$. We have also investigated some quasi-1D solutions of the 2D equation (see Ref. [16]) and have found these solutions to be unstable to transverse perturbations.

In this Letter we have shown that the two-dimensional evolution equation, describing the three-dimensional spatiotemporal evolution of the air-liquid interface of a thin film on the underside of a cooled horizontal plate, has a Liapunov functional that is strictly nonincreasing in time. We use this fact to demonstrate the existence of stable axisymmetric localized patterns for a wide range of parameter values. We have found that for given parameter values in the equation there exists a continuum of possible localized structures (which differ from one another in their total mass) for the amplitude larger than some critical value. The initial conditions that can lead to such states and the transient behavior prior to the formation of the states are still open questions and can only be resolved upon the efficient numerical solution of the full two-dimensional nonlinear evolution equation.

A.O. acknowledges the hospitality of the Institute for Computational Mechanics in Propulsion (ICOMP) at NASA Lewis Research Center in Cleveland where the present work was done. His work is partially supported by the V.P.R. Technion Israel–Mexico Energy Research Fund and Grant-in-Aid of the Government of the State of Israel.

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[16] This assumes that the initial conditions are such that the solution does not settle on a saddle point. For example, a quasi-1D Gaussian solution (localized in only one direction and independent of the other direction) can relax to a quasi-1D localized solution which is unstable to perturbations transverse to the direction of localization. If there is ever any doubt about the stability of a solution, it can always be perturbed with a small amount of noise (with zero mean so as to conserve Q).