RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 41, NUMBER 10 15 MAY 1990

Weakly nonlinear states as propagating fronts in convecting binary mixtures

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(Received 27 November 1989)

We present a picture of the weakly nonlinear time-dependent (blinking) traveling-wave state in the convection of binary mixtures as a propagating, spatially confined solution of coupled Landau-Ginzburg equations. Quantitative agreement with the measured slow oscillation frequency is found. In addition, a number of experimental observations regarding the blinking and confined states can be understood in this picture.

The study of the convection of binary fluid mixtures has proven to be fruitful in furthering our understanding of the spatial and spatio-temporal behavior of a number of driven nonlinear systems that undergo a bifurcation to a traveling-wave type of instability. Examples of such systems are Tollmein-Schlichting waves in channel flows,[1] spiral waves in Couette-Taylor flow,[2] and the secondary bifurcation to the oscillatory instabilities in the convection of pure fluids with low Prandtl number.[3]

In a binary fluid the parameters affecting the flow are the Rayleigh number R, which is the dimensionless temperature difference ΔT, imposed across the fluid layer, and the separation ratio ψ, which is a measure of the coupling between temperature and concentration gradients induced by the Soret effect.[4] It is the sign and value of ψ that determine whether the convection will be oscillatory or stationary. For $\psi < 0$, the competition between temperature and concentration gradients gives rise to a Hopf bifurcation at R_c to traveling waves (TW) in the system.

Experimentally, traveling waves were first observed[5,6] to extend over the entire length of the (rectangular) cell. For cells of sufficiently narrow dimensions, the patterns were observed to be one dimensional in appearance. Later experiments, performed in a closer vicinity of the convective onset, revealed the existence of time independent, spatially confined TW states[7,8] (confined states). Subsequent experiments, performed still nearer to the convective onset[9,10] showed the existence of spatially modulated, time-dependent states (blinking states) where the TW first propagates, for example, to the right and is confined to the right side of the cell, it then fades as a left-propagating TW confined to the left of the cell appears. The cycle then repeats. At certain values of ψ, even closer to the convection threshold, a time independent superposition of spatially-modulated left- and right-going TW (counter propagating waves) has been observed.[9]

Work by Cross[11] has shown that all of the above states exist as solutions of the following coupled Landau-Ginzburg (LG) equations together with finite reflection of left- (A_L) and right- (A_R) going traveling-wave envelopes at the lateral boundaries of the cell

$$\tau_0(\partial_t+s\partial_x)A_R=(1+ic_0)\epsilon A_R+(1+ic_1)\xi_0^2\partial_x^2A_R+g_1(1+ic_2)|A_R|^2A_R+g_2(1+ic_3)|A_L|^2A_R\,,$$
$$\tau_0(\partial_t-s\partial_x)A_L=(1+ic_0)\epsilon A_L+(1+ic_1)\xi_0^2\partial_x^2A_L+g_1(1+ic_2)|A_L|^2A_L+g_2(1+ic_3)|A_R|^2A_L\,. \qquad (1)$$

Here ϵ is the system's control parameter $[\epsilon=(R-R_c)/R_c]$, τ_0 and ξ_0 are the system's characteristic time and length, respectively, s is the TW group velocity, and g_1, g_2, c_i, $i=0,3$ are real parameters. The values of these parameters are ψ dependent. Simulations performed by Cross with all of the c_i taken to be 0, showed that both the qualitative spatio-temporal behavior and order of appearance of the experimentally observed states as a function of ϵ are reproduced by this theory. In addition, on the basis of work done on the transition from "convectively" to "absolutely" unstable states in the framework of a single LG equation,[12] a transition from the spatially modulated, lower lying states to the spatially homogeneous full-cell TW states was predicted to occur at a value of $s^*=2$, where $s^*=s\tau_0/\xi_0\epsilon^{1/2}$ is the dimensionless group velocity. Such a transition was indeed observed to occur experimentally[13] over a broad range of ψ, but at a value of $s^*=1.6$ instead of the predicted value.

Another success of Cross's theory is a complete quantitative description of the dynamical and spatial structure of the linear oscillatory transients observed in a cell of finite lateral size. A result of this model is a shift in the onset of the Hopf bifurcation, $\epsilon_{\rm lin}>0$, resulting from the spatial propagation of the TW. Growth of the instability can only occur in the system for values of $\epsilon > \epsilon_{\rm lin}$ when the losses to the TW amplitude due to imperfect reflection at the lateral boundaries of the cell are overcome by temporal growth, at rate ϵ/τ_0, as the TW traverse the system. Therefore, in an experiment, the observed critical temperature difference of convective onset $\Delta T_{c(\rm expt)}$, will always be higher than the critical temperature difference ΔT_c, for a cell of infinite lateral extent. Recent experiments quantitatively confirm the predictions of the linear theory.[13,14]

On the other hand, our understanding of the behavior of the nonlinear states is not as clear. Experimentally, the following unexplained features have been observed:

(1) The modulation (blinking) period of the blinking state is much larger than any characteristic propagation time of the system (e.g., the time required for a TW to traverse the cell).[9,10] The origin of this time scale is not understood.

(2) For a given value of ψ, the region of existence in R (or ΔT) of the confined and full-cell TW branches is dependent only on ΔT_c and not on the observed critical onset.[13] Although the value of $\Delta T_{c(\rm expt)}$ can be shifted either above or below the lower end of the confined branch by varying the lateral boundary conditions, the behavior of the branch depends solely on $\epsilon = (\Delta T - \Delta T_c)/\Delta T_c$.

(3) In contrast to the above, the blinking states are never observed for values of $\Delta T < \Delta T_{c(\rm expt)}$. In the cases where they have been observed (when the lower end of the confined branch is above $\Delta T_{c(\rm expt)}$) they exhibit negligible hysteresis relative to the experimental onset.[15,16]

The purpose of this work is to introduce a simple explanation for the blinking states based on Eq. (1). The main idea is that these states are essentially propagating-front-like solutions of the coupled LG equations that obey the "marginal stability" theory[17–19] of front propagation. This theory, which has been verified analytically,[19] numerically,[17,18] and experimentally,[20] is relevant to a large class of physical systems which include the complex LG equations. The theory states that the propagation velocity of such nonlinear states is entirely determined by the behavior of the *linearized* equations in the region of small amplitude behind the front. The velocity then selected by the entire state will be the *slowest* for which the front is stable. This approach yields quantitative agreement with the experimentally measured blinking frequencies, together with reasonable explanations for the above experimental observations.

As in the experiments, we will assume that at a given instant a coherent front-type solution of Eq. (1) exists, in which one TW grows from very low amplitudes ($A_R \sim 0$) at one side of the cell to a saturated amplitude ($A_R \gg A_L$) on the other side. The dynamics of the front are then determined by Eq. (1), linearized about $A_R = 0$:

$$\tau_0(\partial_t + s\partial_x)A_R = (1+ic_0)\epsilon A_R + \xi_0^2(1+ic_1)\partial_x^2 A_R + g_2(1+ic_3)|A_L|^2 A_R\,. \qquad (2)$$

Note that we cannot neglect the cross-coupling term since in the region of the front, $A_L \geq A_R$. In this region we can take A_L to be constant[21] (since it is slowly changing in x and t) and of order $\epsilon^{1/2}$ [since only then should Eq. (1) be valid]. Because of the stability of TW over standing waves in our system,[22] $g_2 < 0$ and we have

$$\tau_0(\partial_t + s\partial_x)A_R = \epsilon[(1-a)+i(c_0 - ac_3)]A_R + \xi_0^2(1+ic_1)\partial_x^2 A_R\,, \qquad (3)$$

where $a \equiv -g_2|A_L|^2/\epsilon$ is a term which shows the effective renormalization of ϵ in the vicinity of the front due to the interaction between the left- and right-going TW ($1 > a > 0$).

Equation (3) is solved by substituting $A_R = \exp(kx - \omega t)$ yielding

$$\omega = sk - \frac{\epsilon[(1-a)+i(c_0-ac_3)] + \xi_0^2(1+ic_1)k^2}{\tau_0}\,. \qquad (4)$$

The propagation velocity of the front and therefore of the entire state is given by the envelope velocity $\mathrm{Re}(\omega)/\mathrm{Re}(k)$ of A_R as defined by Eq. (4)

$$\mathrm{Re}(\omega)/\mathrm{Re}(k) = s - \frac{1}{k_r\tau_0}[(1-a)\epsilon + \xi_0^2 k_r^2(1+c_1^2)]\,, \qquad (5)$$

where $k = k_r + ik_i$ and the condition, $\mathrm{Im}(\partial\omega/\partial k) = 0$, for the linear stability of the state was used.

We now need to look at the stability of the front. Following Dee and Langer,[17] we find that such a solution is stable if $\mathrm{Re}(\omega)/\mathrm{Re}(k) \geq \mathrm{Re}(\partial\omega/\partial k)$ since it "outruns" any perturbation to it. This condition gives us a continuum of possible solutions for k_r. We now invoke the "marginal stability" theory[17–19] which states that the system will select the velocity for which $\mathrm{Re}(\omega)/\mathrm{Re}(k) = \mathrm{Re}(\partial\omega/\partial k)$.

Using the marginal stability condition we obtain

$$\mathrm{Re}(\omega)/\mathrm{Re}(k) = \mathrm{Re}(\partial\omega/\partial k) = s - \frac{2(1+c_1^2)^{1/2}(1-a)^{1/2}\epsilon^{1/2}\xi_0}{\tau_0}\,. \qquad (6)$$

From Eq. (6) we now can understand the blinking state as a front-like state which traverses the cell in the same direction but slower than the underlying TW. As this occurs, the downstream end of the state is reflected at the endwalls, thereby forming a left-going state which after passing through (and being suppressed by) the higher amplitude right-going state, will itself begin to grow in space and time, thus forming a left-propagating front whose velocity is also given by Eq. (6). The round trip time of these propagating fronts will be the blinking (or slow modulational) period T of the state

$$T = \frac{2\Gamma}{\partial\omega/\partial k} = \frac{2\Gamma\tau_0}{s\tau_0 - 2(1-a)(1+c_1^2)^{1/2}\xi_0\epsilon^{1/2}}\,, \qquad (7)$$

where Γ is the aspect ratio of the cell. In Eq. (7), the parameters τ_0, ξ_0, c_1, and s have either been measured[14,16] or calculated.[23] The exact value of a cannot be estimated theoretically since g_2 and $|A_L|^2$ are not known. On the other hand, we know that a negative value of the front propagation velocity would correspond to the transition from the spatially modulated "convectively unstable" states to the spatially homogeneous "absolutely unstable" states of the system. As previously mentioned, this transition was experimentally observed to occur at a value of $s^*=1.6$. By setting $\partial\omega/\partial k=0$ in Eq. (6), we find the predicted value for s^* at the transition to be $2(1-\alpha)^{1/2}(1+c_1^2)^{1/2}$. Using this determination of α, we now have both a plausible explanation for the occurrence of the transition from convective to absolute instability at a value of $s^*<2$, and a prediction for the blinking frequency having *no* adjustable parameters.

In Fig. 1 we compare measurements of the blinking-state period as a function of ϵ at $\psi=-0.02$ for two cells of different Γ with the periods predicted by Eq. (7). We used the measured values of $\tau_0=0.105\tau_v$ and $s=0.93v_p$ and the calculated one[23] of $\xi_0=0.381d$. Here, d is the cell height, τ_v the vertical thermal diffusion time, and v_p the TW phase velocity at the onset of convection. The agreement with the predictions is seen to be quite good.

Given the picture of the blinking state as a propagating envelope function that is imperfectly reflected at the cell's endwalls, we can understand the dependence of the stability of the blinking branch on $\Delta T_{c(\text{expt})}$. As in the case of the linear state, the blinking state must compensate for reflective losses by growth as it traverses the cell to remain stable. Thus, we would expect the lower end of the blinking state branch to appear approximately at the same point as $\Delta T_{c(\text{expt})}$ since the two states exhibit similar growth at small values of ϵ.

In contrast to the blinking states, the confined TW branch would not be expected to relate to $\Delta T_{c(\text{expt})}$ in this picture. The envelope function of the confined branch is stationary in time and, is therefore, not reflected at the boundaries. We would like to note that Eq. (7) predicts the existence of a confined state at a *single* value of ϵ where $\text{Re}(\partial\omega/\partial k)=0$. Although this value of ϵ does correspond to the point where the observed confined branch loses stability to the full-cell TW branch, neither the existence of the confined state over a *range* of ϵ nor the aperiodic behavior of the blinking state observed for (higher) values of ϵ immediately preceding the confined branch are explained by this mechanism.

In conclusion, we have introduced a simple picture of the blinking state as a continuously propagating front-like solution of the coupled Landau-Ginzburg equations introduced by Cross. Using a marginal stability analysis to calculate the propagation velocity of the leading edge of such a solution, we were able to provide good quantitative agreement with experimental measurements of the modulational period of the state. In addition, the qualitative picture of a spatially confined propagating solution coupled with reflection at the lateral boundaries of the system provides plausible explanations for some recent experimental observations.

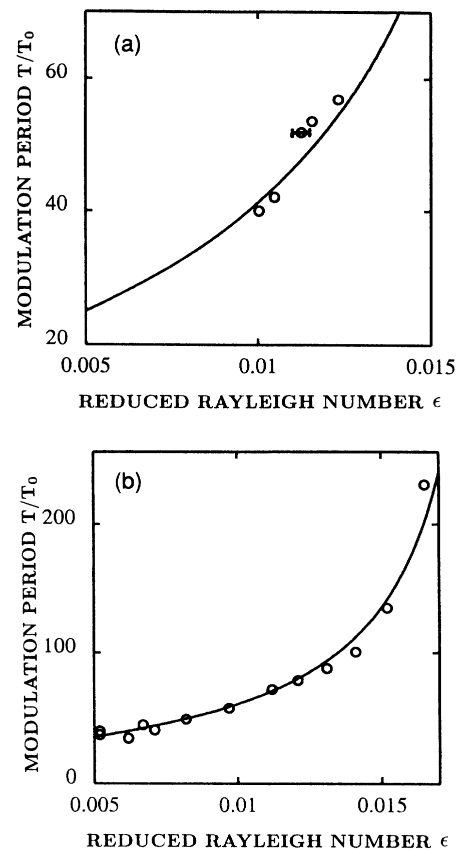

FIG. 1. Blinking (modulation) period, T normalized by the oscillation period T_0 of the linear TW state as a function of ϵ. The solid lines in both (a) and (b) are the theoretical predictions given by Eq. (7). (a) $\psi=-0.022$ in a 27.5 wt.% ethanol-water mixture with $\Gamma=12.0$ and an average temperature of 29.05 °C across the cell. The error bar shown is representative and corresponds to a 1-mK uncertainty in the temperature difference. (b) $\psi=-0.021$ in a 0.3 wt.% ethanol-water mixture with $\Gamma=16.25$ and an average temperature of 21.43 °C across the cell.

One of us (P.K.) acknowledges the collaboration of C. M. Surko. J.F. acknowledges support by the Department of Energy Office of Basic Energy Sciences.

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