

Nonperiodic time dependence at the onset of convection in a binary liquid mixture

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We report measurements of the convective heat transport near the onset of convection in a normal ^3He - ^4He mixture for values of the separation ratio ψ between -0.044 and -0.010 . For $\psi \lesssim -0.018$, the first bifurcation was to a relatively large-amplitude time-dependent state with characteristic frequencies over an order of magnitude smaller than predicted by linear-stability analysis. The onset of the time dependence was intermittent and not hysteretic. Above onset, the frequency spectrum of the time dependence either contained discrete frequencies or not, depending on ψ .

Convection in shallow, horizontal layers of binary fluid mixtures heated from below has attracted much attention recently because it offers the opportunity to study a great variety of linear and nonlinear phenomena.^{1,2} In this system there are two control parameters. One of them, the Rayleigh number R , is proportional to the vertical temperature difference ΔT across the fluid layer. The other, the separation ratio ψ , determines whether concentration gradients help ($\psi > 0$) or hinder ($\psi < 0$) convection. By changing ψ , the nature of the bifurcation from the conducting to the convecting state, which occurs when R is increased beyond $R_c(\psi)$, may be altered. In this communication we are concerned with the convective heat transport in the region $\psi < \psi_c = -0.010$. Close to but below ψ_c convective motion is initiated in our system via a backward Hopf bifurcation to a time-periodic state^{3,4} which presumably consists of traveling waves.⁵⁻⁸ The characteristic frequency ω of this state differs from the Hopf bifurcation frequency⁹⁻¹¹ by only a factor of 0.6 or so, and when scaled by the vertical thermal diffusion time t_v is approximately equal to 2. We found that for $\psi < \psi_i \cong -0.018$ the first bifurcation changes dramatically. The convecting state just beyond R_c has an intermittent character, apparently forming and disappearing in bursts with a duration which is much shorter than the irregular separation between them. The convective heat transport between bursts is zero within our resolution. Separation between bursts of several hundred times T_v have been observed. The intermittent character of this state suggests a relationship to the noise-induced convectively (as opposed to absolutely) unstable solutions of the complex Ginzburg-Landau equation (CGLE) which have been studied recently.^{12,13} As R is increased, the time between bursts becomes shorter; and for sufficiently large R successive ones begin to overlap. The spectrum of the convective heat transport can have periodic components or be broadband, depending on R and ψ .

Over a wider range of ψ but in less detail than explored quantitatively by us, the onset of time dependence has been observed previously;¹⁴ but no characterization of the bifurcation lines and of the nonlinear state have been attempted.

Our convection cell had upper and lower boundaries of copper and sidewalls made of thin stainless steel. The cell

was a rectangular box with vertical height $d = 0.083$ cm, length $34d$, and width $6.9d$. The temperature of the top plate was held constant to ± 0.5 μK . Power was dissipated in a resistance heater on the bottom plate and the bottom plate temperature was monitored as a function of time by a germanium thermometer. We thus had the ability to measure the temperature difference across the cell, the average temperature in the fluid, and the (time-dependent) thermal conductance of the fluid layer. Since we are interested in the convective part of the heat transfer, we express our results in terms of $N - 1$, where the Nusselt number N is the measured conductance normalized by the conductance of the nonconvecting state. All times and frequencies are normalized by the vertical thermal diffusion time $t_v = d^2/\kappa \approx 24$ sec (κ is the thermal diffusivity). The fluid had the same 0.03 molar concentration of ^3He in ^4He used in two previous experiments^{3,15,16} and the relationship between ψ and the average temperature used here is the same as in the previous work. The Prandtl number $\sigma = \nu/\kappa$ was about 0.6, and the Lewis number $L = D/\kappa$ was close to 0.03 (ν is the kinematic viscosity and D the mass diffusivity). The temperature was always maintained above the superfluid transition temperature for the mixture.

Figure 1 is a bifurcation diagram for the parameter region we have studied. For $\psi_i \lesssim \psi \lesssim \psi_c$, the bifurcation sequence for increasing ΔT is as follows:³ At the open squares, pure conduction lost stability at a Hopf bifurcation to a state we will call the periodic state. The periodic state was characterized by a mean convective heat transport $N - 1 \approx 10^{-4}$ with periodic modulations $\approx 10^{-5}$ at a modulation frequency $\omega \approx 2$. These observations are consistent with the periodic state being composed of traveling waves (TW) with a time-independent envelope and finite spatial extent.^{7,8} The bifurcation to the periodic state was hysteretic at these values of ψ , and ΔT had to be reduced to the open diamonds before pure conduction was recovered. As ΔT was raised above the onset (ΔT_c) of the periodic state, the modulation frequency decreased linearly with a slope $\partial\omega/\partial\varepsilon \approx -18$, where $\varepsilon = (\Delta T - \Delta T_c)/\Delta T_c$. At the open triangles, the periodic state lost stability to a much larger amplitude ($N - 1 \approx 10^{-2}$), but time-independent state that we refer to as stationary convection. This bifurcation was also hysteretic, and the values of ΔT

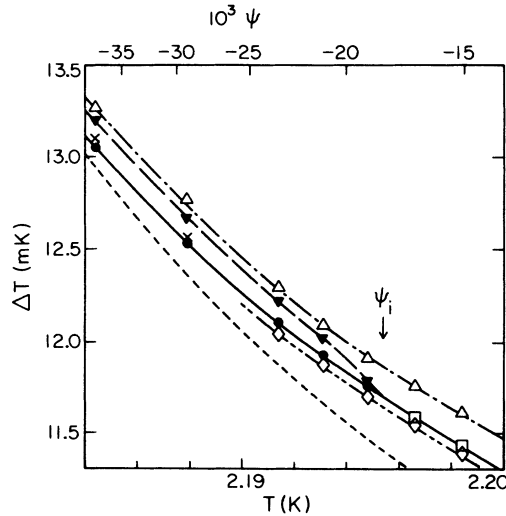


FIG. 1. Bifurcation diagram showing lines to transition between various states. Open squares: conduction to periodic oscillations. Open triangles: periodic oscillations to steady convection. Short dashed line: steady convection to conduction. Open diamonds: periodic oscillation to conduction. Solid circles: conduction to the slow state. Solid triangles: slow state to periodic oscillations. Crosses: periodic oscillations to the slow state. Points are experimentally determined transitions, lines are a guide to the eye.

at which pure conduction reappears is shown as the short dashed line in the figure. The periodic state is described in more detail elsewhere in connection with its behavior closer to the codimension two point.³

Of central interest in this communication is the existence of a state for $\psi \leq \psi_i$ which has a much slower time dependence than the periodic state described above. An interesting characteristic of the slow state is that it is the first convecting state and yet its onset is intermittent. As shown in Fig. 2 for $\psi = -0.021$ and $\epsilon = (0.3 \pm 0.3) \times 10^{-3}$, the convective heat transport just above onset consists of irregularly spaced bursts. Note that the time between bursts is very long compared to t_v . The bifurcation to the slow state is shown in Fig. 1 as solid circles and appears along a line that is a smooth extension of the bifurcation line to the periodic state (open squares). The bifurcation to the slow state is not hysteretic. The amplitude variations of the Nusselt number are much larger in the slow state than in the periodic state and the charac-

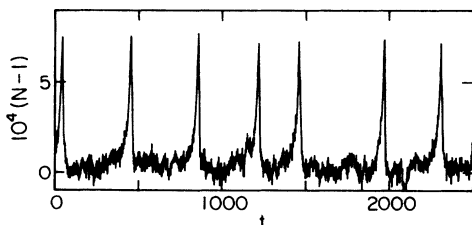


FIG. 2. Convective heat-transport time series for $\psi = -0.021$, $\epsilon = (0.3 \pm 0.3) \times 10^{-3}$.

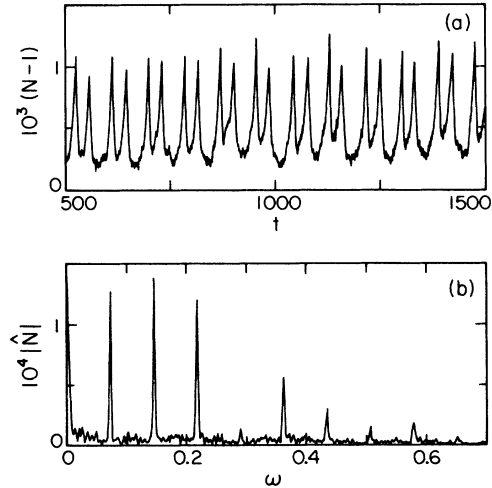


FIG. 3. (a) A section of a $3000t_v$ convective heat-transport time series for $\psi = -0.021$, $\epsilon = (3.6 \pm 0.3) \times 10^{-3}$. (b) Fourier transform of the complete time series.

teristic frequency is more than an order of magnitude smaller. The slow state is a stable state of the system which we have observed under stationary external conditions for as long as $10^4 t_v$.

At any value of ψ , the characteristic frequency of the slow state increases with ϵ . The nature of the time dependence varies with ψ . Figure 3 shows a portion of a convected heat-current time series and the Fourier transform of the entire $3000t_v$ time series for $\psi = -0.021$ and $\epsilon = (3.6 \pm 0.3) \times 10^{-3}$. [Note the large change in time scale between Figs. 2 and 3(a)]. The Fourier transform reveals a single frequency ($\omega = 0.073$) with a rich harmonic structure and a broadband contribution that is about two times larger than the instrumental background. By contrast, Fig. 4 shows a portion of the convective heat transport time series for $\psi = -0.044$ and $\epsilon = (7.6 \pm 0.5)$

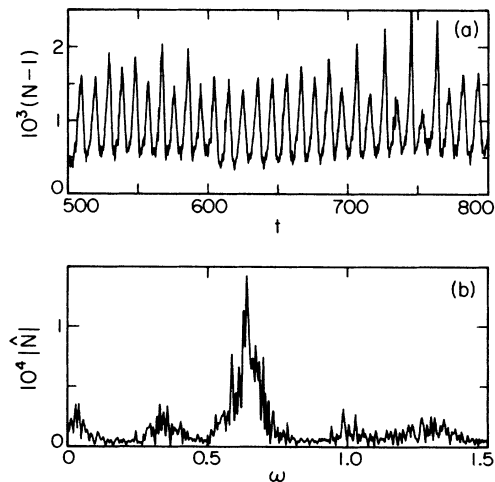


FIG. 4. (a) A section of a $3000t_v$ convective heat-transport time series for $\psi = -0.044$, $\epsilon = (7.6 \pm 0.5) \times 10^{-3}$. (b) Fourier transform of the complete time series.

$\times 10^{-3}$ and the Fourier transform of the complete time series. The spectral resolution is $\Delta\omega = 4.2 \times 10^{-3}$. Successive points in the spectrum scatter randomly about a smooth curve, showing that at this value of ψ the Fourier transform has mostly broad features. We have examined Fourier transforms of all three time series and find no evidence for a periodic component at frequencies corresponding to the TW state.

Another characteristic of the slow state in this range of ψ is that as ΔT is raised to the values labeled by the solid triangles in Fig. 1, a hysteretic bifurcation occurs in which the slow state dies out and a stable periodic state remains. This state has all the same characteristics of the TW's which form via the backward Hopf bifurcation for $\psi > \psi_i$. If ΔT is raised further, the periodic state loses stability to stationary convection at the open triangles, but if ΔT is decreased, one of two events will occur, depending on ψ . For $\psi \gtrsim -0.025$, the periodic state remains stable to the points labeled by open diamonds, below which pure conduction is restored. For $\psi \lesssim -0.025$, the periodic state remains stable only until reaching the crosses, where it loses stability to the slow state.

To more clearly illustrate the relationship between the slow state and the periodic state, the following transient measurement was made: At $t=0$, with $\psi = -0.038$, ε was changed from $-(1.4 \pm 0.3) \times 10^{-3}$ to $(11.2 \pm 0.3) \times 10^{-3}$, i.e., from conduction to a point where the slow state is unstable to the periodic state. The resulting time series is shown in Fig. 5. After $20t_v$, the slow state, with its characteristic large amplitude and slow oscillations, grew out of the conducting state. After $170t_v$, the slow state decayed to the periodic state. A Fourier transform is required to resolve the time dependence of the periodic state, but here its mean conductance can be compared with that of the conductive state and the amplitude variations of the slow state.

At sufficiently small amplitudes, traveling waves in binary mixtures should have an envelope that satisfies the complex Ginzburg-Landau equation (CGLE).¹⁷ A number of recent theoretical papers have described the interesting dynamics of this generic amplitude equation^{12,17-19} and its application to binary mixtures.^{13,17,19-21} Our periodic state has the characteristics of a traveling-wave state with a time-independent envelope. One would hope that our results for the slow state can also be understood on the basis of the CGLE, without invoking instabilities beyond the scope of that equation. We presume that in the region of slow time dependence this envelope has become unstable and that the time dependence is due to this envelope instability. We note that the bursts in Figs. 2-5 reach values of $N=1$ which

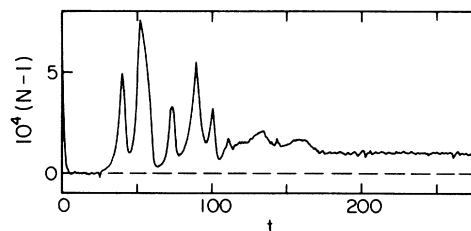


FIG. 5. Convective heat transport as a function of time showing transitions between conduction, the slow state, and the periodic state for $\psi = -0.038$. At $t=0$, ε was changed from $-(1.4 \pm 0.3) \times 10^{-3}$ to $(11.2 \pm 0.3) \times 10^{-3}$. (A Fourier transform is required to resolve the periodic oscillations for $t > 180$).

are an order of magnitude larger than the time-averaged value of $N-1$ in the periodic state (see Fig. 5). This suggests that the oscillations first form throughout a large portion or all of the cell, and in the intermittent cases (Fig. 2) disappear altogether between the bursts. If this is indeed the case, then the periodic state would seem to be highly localized,^{7,8} occupying only about 10% or so of the cell.

There are a number of mechanisms which can produce traveling-wave states with time-dependent envelopes, but as yet no complete explanation of our results. Deissler and Brand^{12,13} have shown that a generic feature of the CGLE with a nonzero group velocity is a finite range of $\varepsilon > 0$ where the system is convectively unstable and thus sensitive to spatially varying sources of noise. Also in the presence of noise the CGLE can display subcritical ($\varepsilon < 0$) bursts.^{13,22} Either of these might provide a natural explanation of the observed intermittency at the onset of the slow state. Cross²¹ has demonstrated a mechanism for producing slow, periodic modulations of a traveling-wave state in the absence of noise. However, it is not clear if his mechanism can explain other features of the slow state, such as its nonperiodic behavior, the ε dependence of its characteristic frequency, and the hysteretic transition to the periodic state. A direct comparison with theory awaits either a theoretical calculation or experimental measurement of the nonlinear parameters in the CGLE. A determination of these parameters is also important for deciding whether the Benjamin-Feir instability¹⁷ plays a role.

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¹For a review of the older literature, see J. K. Platten and J. C. Legros, *Convection in Liquids* (Springer, New York, 1984), Chap. 9.

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