

Convection in a Binary Mixture Heated from Below

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Heat-transport measurements in a bulk normal-fluid ^3He - ^4He mixture heated from below and over the range $-0.02 \leq \psi \leq 0.02$ of the separation ratio ψ reveal a forward bifurcation with an initial slope $S \cong 0$ of the Nusselt number for large ψ , and a backward bifurcation for $\psi < \psi_b \cong 0.006$. At ψ_b , $S \cong 1$. The critical line $\Delta T_c(\psi)$ has two branches which meet at $\psi \cong -0.003$, and which we attribute to the expected stationary and Hopf bifurcation lines. However, stable oscillations bifurcating from the conduction state exist only for $\psi \leq -0.015$.

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In a horizontal layer of fluid heated from below, a transition, or bifurcation, occurs from conduction to convection when the temperature difference ΔT reaches ΔT_c . Depending on the values of relevant externally controlled parameters, the fluid velocity may grow continuously as ΔT increases beyond ΔT_c , or it may jump precipitously to a finite value. We refer to the former case as a forward, and to the latter as a backward bifurcation. In analogy to equilibrium phase transitions, we shall call the marginal case between them a tricritical bifurcation. After a backward bifurcation, fluid flow will persist even when ΔT is reduced below ΔT_c , until a so-called saddle node is reached where convection ceases abruptly. We call a transition a Hopf bifurcation or a stationary bifurcation, depending on whether the velocity of the convecting state is time-periodic or constant. It may occur that a line of Hopf bifurcations in parameter space meets a line of stationary bifurcations. Such a point is a codimension-two bifurcation. Convection in liquid mixtures has provoked considerable attention recently because it reveals a number of these interesting nonlinear phenomena. They are associated with the interaction between heat diffusion and mass diffusion.¹⁻⁷ Theoretical investigations have been limited largely to the physically unrealistic case of permeable and slip top and bottom boundaries and thus are not expected to be *quantitatively* reliable. We present in this paper the results of an experimental investigation which reveal that even the qualitative features predicted by the theory are not all shared by the physical system with rigid, impermeable boundaries.

In the case of binary mixtures there are *two* parameters which can be controlled externally. One of them, the Rayleigh number R , is proportional to the imposed temperature difference ΔT . The other, the separation ratio ψ , is proportional to the thermodiffusion ratio k_T and can in our case be varied by changing the mean operating temperature. Thus, a wide range of the R - ψ plane can be explored experimentally. For $\psi > 0$ ($\psi < 0$) concentration gradients will tend to enhance (suppress) convection. The theoretical prediction for the phase diagram is illustrated in Fig. 1(a). Albeit in part from calculations with unphysical boundary condi-

tions, one expects the following. The convecting state for values of R slightly greater than the critical value R_c will be time-periodic or stationary, depending on the value of ψ .^{1,2} For sufficiently negative ψ , there is a Hopf bifurcation at $R_{co}(\psi)$ (dashed line). For larger ψ , the bifurcation at $R_{cs}(\psi)$ leads to steady convection (solid line). The two bifurcation lines meet in the R - ψ plane at the codimension-two (CT) point when $\psi = \psi_{pc} < 0$.^{1,3} At that point, the stationary bifurcation is backwards.^{1,3} It is predicted to become forward *via* a tricritical bifurcation when ψ is increased to ψ_t .³ For positive ψ , R_{cs} is reduced and the initial slope of the heat-transport (Nusselt number) curve is predicted to be dramatically depressed.

The experimental results are summarized in part in Fig. 1(b). Over the range $-0.02 \leq \psi \leq 0.02$ we find

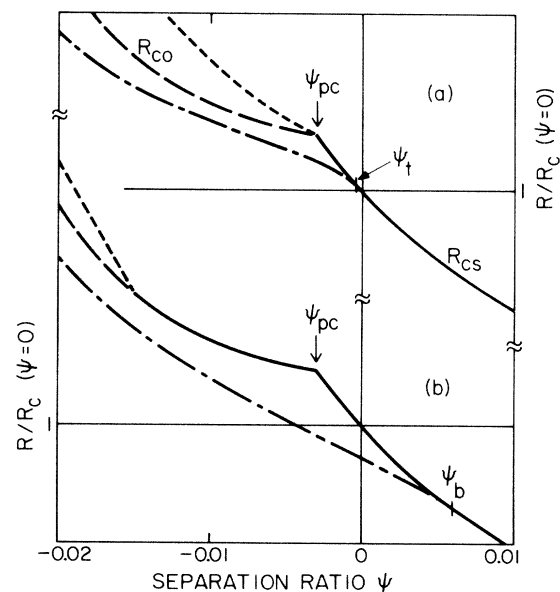


FIG. 1. (a) Theoretical and (b) experimental phase diagram in the R - ψ plane. The vertical scale is schematic except for the point $R/R_c(\psi=0)=1$. Solid lines, stationary bifurcation at $R_{cs}(\psi)$; dash-dotted lines, saddle-node bifurcation; long dashed lines, bifurcation from pure conduction to oscillations at $R_{co}(\psi)$; short dashed lines, bifurcation from oscillations to a convecting state (heteroclinic orbit in the theory).

the following features. (1) For sufficiently large positive ψ , there is a forward bifurcation to steady convection at a small value of R_{cs} , and the initial slope S of the Nusselt number is very small or zero. These observations are consistent with the theory. (2) When ψ is reduced below $\psi_b \approx 0.006$, the stationary bifurcation becomes hysteretic. The theory predicts a change from a forward to a backward bifurcation *via* a tricritical bifurcation at $\psi = \psi_t < 0$,³ whereas we find $\psi_b > 0$. The saddle node which results from the backward bifurcation is shown as dash-dotted lines in Fig. 1. (3) For $\psi < \psi_b$ there exist two convecting states, one at relatively small and the other at larger R [this is not shown in Fig. 1(b)]. The corresponding heat-transport curves are qualitatively similar to those encountered in non-Boussinesq systems⁸ where there are separate ranges of stability for three-dimensional (hexagonal) and two-dimensional (roll) flow. The existence of two states has not yet been predicted. (4) For $-0.015 \leq \psi < \psi_{pc}$, we found no Hopf bifurcation to a time-periodic state from the conduction state. The theory predicts a forward Hopf bifurcation at R_{c0} for this range of ψ . Stable oscillations about the conduction state were found only for $\psi \leq -0.015$. (5) The curve $R_c(\psi)$ for the backward bifurcation has two branches which meet at a value of ψ rather close to that expected for ψ_{pc} . We conjecture that there exists a Hopf bifurcation for $-0.015 \leq \psi < \psi_{pc}$ which immediately triggers a transition to the convecting branch associated with the backward *stationary* bifurcation. (6) The convecting state which exists at small R and for $\psi < \psi_b$ is always a stationary flow. The branch existing at larger R has a *very* slow periodic time dependence (periods up to 400 vertical thermal diffusion times) for some of the parameter values used in our work. There are no predictions for the time dependence of the convecting state.

The convection cell was installed in an apparatus used previously.⁶ It was rectangular, had a height d of 0.110 cm, and aspect ratios $L_x = l_x/d = 26.0$ and $L_y = l_y/d = 6.50$ (l_x and l_y are the length and width of the cell). The fluid was the *same* ^3He - ^4He mixture that had been used for the study of the codimension-two bifurcation in a porous medium.^{6,9} We are therefore able to use the relationship $\psi(T)$ established from that work.¹⁰ With increasing T , ψ increases from negative values and passes through zero at $T = 2.223$ K.⁶ The Lewis number is close to 0.03. The Prandtl number σ is 0.6.

Since we cannot observe the pattern in the liquid-helium experiment, we made shadowgraph flow visualizations and Nusselt-number measurements at room temperature using water ($\sigma \approx 6$) and a cell with $d = 0.279$ cm and the same $L_x = 26.0$ and $L_y = 6.50$. From the Nusselt-number measurements as a function of $\epsilon \equiv R/R_c - 1$ we found $S = (dN/d\epsilon)_{\epsilon=0} = 0.90$.

The pattern consisted of thirteen pairs of straight rolls parallel to the short side. It was stable until a transition to twelve pairs occurred with increasing ϵ , near $\epsilon = 3.5$, which we presume to be due to the skewed-varicose instability.¹¹ In the helium experiments, $N(\epsilon)$ evolved smoothly with increasing ϵ for the mixture at all values of ψ (except for the hysteresis near R_c) as well as for pure ^4He . A transition occurred near $\epsilon \approx 0.7$, roughly where we expect the skewed-varicose instability to occur for $\sigma \approx 0.6$. Thus the thermal behavior of the low-temperature system is also consistent with a parallel-roll pattern.

In Fig. 2(b) we show as solid circles the convective

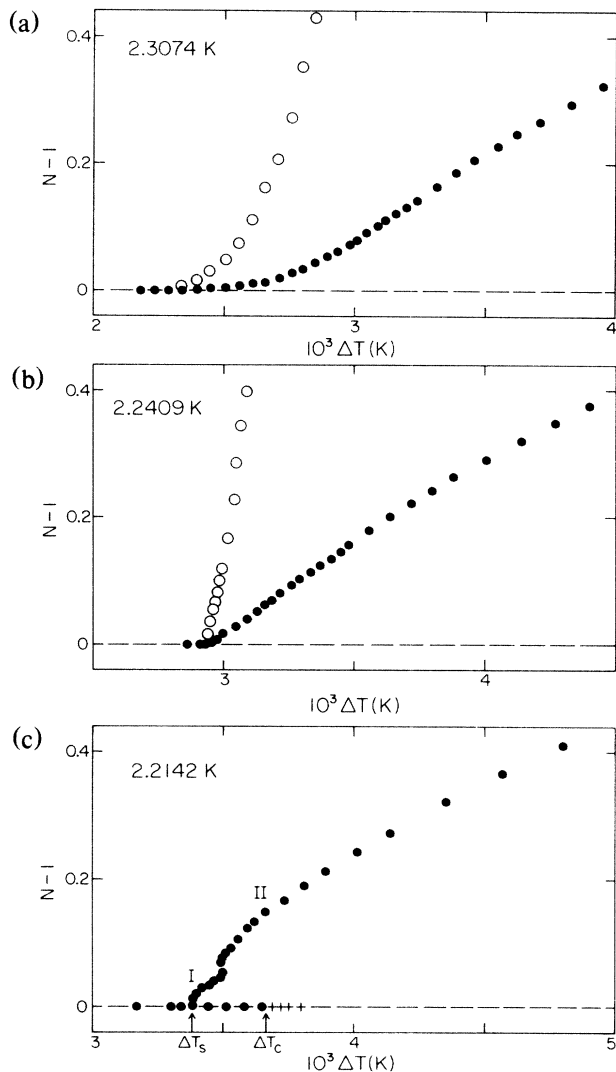


FIG. 2. Convective contribution $N-1$ to the Nusselt number N as a function of the temperature difference across the cell. Open circles are for $10(N-1)$. The indicated temperatures are for the cold end of the cell. Approximate values of the separation ratio ψ are (a) 0.02, (b) 0.006, and (c) -0.004. In (c), the plusses correspond to unstable pure conduction states which decayed to the convecting branch.

contribution $N-1$ to the Nusselt number N as a function of the temperature difference across the cell at a cold-end temperature of 2.2409 K ($\psi \approx 0.006$). There is a sharp nonhysteretic (forward) bifurcation near $\Delta T_c = 2.97$ mK. The sharpness of the bifurcation is illustrated more dramatically by the open circles, which correspond to $10(N-1)$. The initial slope S of N is 1.01, very close to that measured with pure ^4He and with water.

In Fig. 2(a), data similar to those in Fig. 2(b) are shown for $T = 2.3074$ K ($\psi \approx 0.02$). The nature of the heat-transport curve has been altered greatly. The bifurcation occurs at a significantly smaller value of ΔT ($\Delta T_c \approx 2.3$ mK or less), corresponding to a smaller R_c . Near the bifurcation the slope of N is dramatically reduced, by a factor of 20 or more compared to Fig. 2(b). The data are also consistent for instance with $(N-1) \sim \epsilon^2$ near $\epsilon = 0$, corresponding to $S = 0$. The behavior illustrated here for positive ψ is consistent with the theory.^{1,2}

In Fig. 2(c), $N-1$ is shown as a function of ΔT for 2.2142 K ($\psi \approx -0.004$). The bifurcation is clearly hysteretic, with $\Delta T_c = 3.67$ mK and a saddle-node bi-

furcation point at $\Delta T_s = 3.36$ mK. The convecting state has two branches, labeled I and II, one extending from ΔT_s to $\Delta T \approx 3.50$ mK and the other existing from $\Delta T \approx 3.48$ mK up to the skewed-varicose instability near $\Delta T \approx 5$ mK.

Details of the heat-transport curves near the bifurcations for $T = 2.2409$ and 2.2142 K are shown in Figs. 3(a) and 3(b). Here the open (solid) circles correspond to stable states reached by increasing (decreasing) the heat current, and plusses correspond to unstable pure conduction states. The arrows indicate transitions which, at constant heat current, will involve a decrease (increase) in ΔT when N increases (decreases). As can be seen from Fig. 3(a), there is no hysteresis for $T = 2.2409$ K ($\psi \approx 0.006$). Figure 3(b) illustrates in detail the hysteretic transition (backward bifurcation) which occurs for $\psi \leq 0.006$. The existence of the two convecting states I and II is also clearly demonstrated by these data.

In Fig. 4 we show ΔT_s (open circles) and ΔT_c (solid circles) as a function of T . Along the upper edge we give our estimate of $\psi(T)$. We find that ΔT_s varies smoothly with T (or ψ). On the other hand, the experimental values of ΔT_c clearly fall on two branches, labeled 1 and 2, which meet at $T = 2.216$ K or

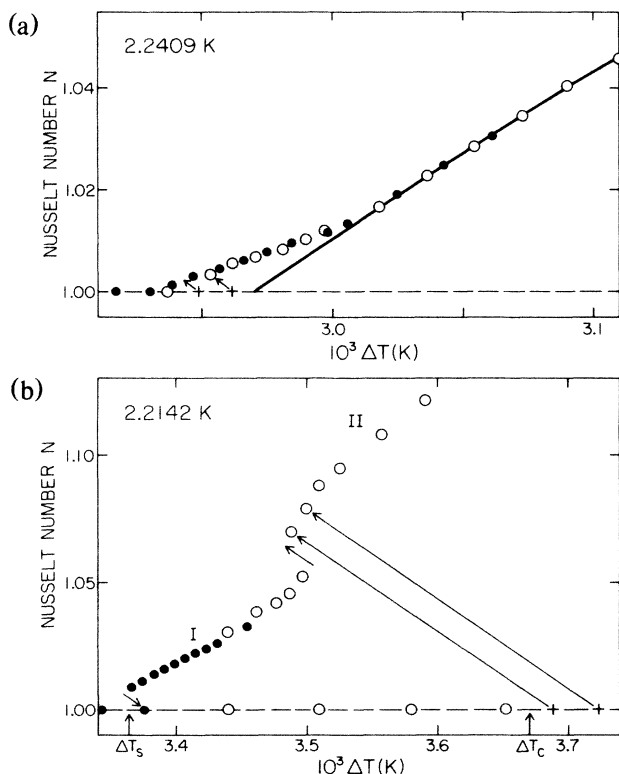


FIG. 3. Expanded view of the Nusselt number as a function of ΔT for two top temperatures, corresponding to (a) $\psi \approx 0.006$ and (b) $\psi \approx -0.004$. Open circles, increasing heat current; solid circles, decreasing heat current. The plusses are unstable pure conduction states which decayed to the convecting branch as indicated by the arrows.

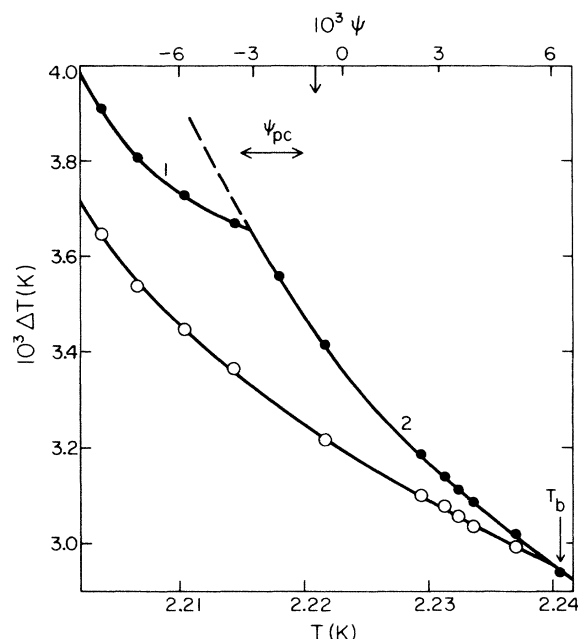


FIG. 4. Open circles, the temperature difference ΔT_s of the saddle-node point [see Fig. 3(b)]; solid circles, the critical temperature difference ΔT_c at the onset of convection with increasing heat current [see Fig. 3(b)]. The values of the separation ratio ψ are given along the top border of the figure. The vertical arrow near the top border corresponds to the codimension-two point ψ_{pc} for the porous medium case. The horizontal double-headed arrow gives the expected location of ψ_{pc} for the bulk mixture.

$\psi \cong -0.003$. The high-temperature branch (2) meets the saddle-node line at $T_b = 2.240$ K, corresponding to $\psi_b \cong 0.006$. The theory³ predicts a negative ψ_t at a *tricritical* point with $(N-1) \sim \epsilon^{1/2}$ for the change from a forward to a backward bifurcation. However, for our Lewis number the departure of $N(\epsilon)$ from nearly linear behavior is expected to be confined essentially to $\epsilon \leq 10^{-3}$ and thus would be difficult to observe.¹² The Nusselt number data shown in Figs. 2(b) and 3(a) correspond very closely to ψ_b . They give an initial slope S of N essentially equal to that obtained with pure ^4He and thus, as expected, do not reveal tricritical behavior. However, the positive ψ_b differs from the predicted negative ψ_t . Our data differ from an experimental result obtained with ^3He - ^4He mixtures by Gao and Behringer,¹³ who report tricritical behavior with a divergent initial slope S of N . Since their Lewis number is similar to ours, we are unable to explain their data.

In Fig. 4, the downward-pointing arrow near $\psi \cong -0.001$ indicates the experimentally observed CT bifurcation in the porous medium.^{6,9} In the bulk fluid, one expects this bifurcation at a somewhat more negative value of ψ .^{1,3} The expected location is dependent upon the value of the Lewis number (which is not known very well) and of σ . It falls somewhere within the range indicated by the double-ended horizontal arrow in Fig. 4, and is thus consistent with the experimentally observed meeting point of the two branches of $\Delta T_c(\psi)$. We believe that the high-temperature branch (2) of $\Delta T_c(\psi)$ is the stationary bifurcation at ΔT_{cs} . We conjecture that the low-temperature branch (1) corresponds to the Hopf bifurcation at ΔT_{c0} . In that case the meeting point at $\psi \cong -0.003$ corresponds to the CT point at ψ_{pc} . However, we have observed no oscillations in its vicinity.¹⁴ Thus, many of the interesting phenomena expected to occur there^{3,6} cannot be studied in this system.

We did observe stable oscillations bifurcating from the conduction state for $\psi \leq -0.015$ [see Fig. 1(b)]. Within our resolution this bifurcation was nonhysteretic. However, at the bifurcation point, the frequency was small and the oscillation amplitude finite.¹⁵ The amplitude at ΔT_{c0} apparently grew to sufficient size to reach the attractor basin of the stationary convecting state when $-0.015 \leq \psi \leq \psi_{pc}$, thus preventing

the observation of stable oscillations over that range.

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¹⁰The relationship $\psi(T)$ is known only approximately. It depends in a sensitive way upon the identification of the temperature where the experimentally observed Hopf frequency of Ref. 6 vanishes with the codimension-two point of Ref. 3. We assumed $\psi(T) = 0$ just above that temperature.

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¹⁵For $\sigma \leq 1$ and certain ranges of the Lewis number and ψ , a backward Hopf bifurcation was obtained theoretically from a Lorenz truncation by M. G. Velarde and J. C. Antoranz, *Phys. Lett.* **80A**, 220 (1980).