Experimental Observation of a Codimension-Two Bifurcation in a Binary Fluid Mixture

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Heat-transport measurements in a normal-fluid ^3He-^4He mixture contained in a porous medium and heated from below show a bifurcation to steady or oscillatory flow, depending on the mean temperature. With increasing Rayleigh number R the oscillatory state is entered via a forward Hopf bifurcation with frequency $\omega_c > 0$. As the stationary bifurcation is approached by change of the mean temperature, ω_c vanishes. With increasing R, the oscillatory state terminates with vanishing frequency and finite amplitude in a hysteretic bifurcation to a steady state. Those observations agree with recent theoretical predictions.

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In a binary fluid mixture contained in a porous medium and heated from below, the convecting state may be either time periodic or stationary, depending on the value of the separation ratio ψ.[1] Thus, there are two bifurcation lines in parameter space; and a codimension-two (CT) bifurcation occurs at the point where these lines intersect. In the vicinity of the intersection, competition between the two nonlinear states leads to interesting linear and nonlinear phenomena. For that reason, CT bifurcations have attracted considerable attention recently.[2] They are also of great current interest because one expects that external modulation of the control parameter by a *single* frequency can lead to chaotic behavior in the immediate vicinity of the convective threshold.[3]

We present sensitive heat-flow measurements at cryogenic temperatures, using a liquid mixture of ^3He and ^4He in a porous medium. We are able to vary ψ through the CT value by changing the mean operating temperature. Our measurements confirm five key theoretical predictions based on an amplitude equation[4] for the region near the CT point, namely: (i) Near the CT bifurcation, the Hopf bifurcation to the time-periodic state is forward. (ii) When ψ is changed so as to approach the CT bifurcation, the frequency of the Hopf bifurcation vanishes. (iii) As the Rayleigh number R is increased beyond the Hopf bifurcation, the frequency goes to zero while the amplitude stays finite. (iv) The second bifurcation (iii above) is hysteretic. (v) The range of R over which oscillations are observed vanishes at the CT point.

The experiments were done with a mixture of molar concentration $X = 0.030$ of ^3He in ^4He at temperatures above the superfluid-transition temperature $T_\lambda = 2.127$ K. This fluid is versatile for reaching the CT point because ψ can be changed over a wide range by a change in the temperature. The separation ratio is given by

$$\psi = -k_T\beta_2/T\beta_1, \tag{1}$$

where $\beta_2 = -\rho^{-1}(\partial\rho/\partial X)_{P,T}$ and $\beta_1 = -\rho^{-1}(\partial\rho/\partial T)_{P,X}$, and where k_T is the thermal diffusion ratio. For $T \cong T_\lambda + 0.014$ K, β_1 vanishes and thus $\psi = -\infty$. At a somewhat higher temperature, k_T changes sign[5] and $\psi = 0$. Therefore the separation ratio can be varied from $-\infty$ to positive values by an increase in the mean temperature. Our measurements cover the range $-0.16 \lesssim \psi \lesssim 0.1$.[6] The Lewis number L (the ratio of the mass diffusivity to the thermal diffusivity) was about 0.03.[5]

The convection cell was 4.76 mm high, 19.18 mm wide, and 38.51 mm long. The top and bottom plates were made of copper which has a high thermal conductivity, and the sidewalls of the low-conductivity material Vespel SP22. The cell contained a porous medium consisting of 1082 nylon spheres with diameter 1.59 mm packed in a body-centered cubic array. Thus, the medium consisted of four layers, containing alternately 12×24 and 11×23 spheres. The cell was filled with the mixture at $T = 2.13$ K at a pressure of 0.2 bar, and sealed by a valve located on top of the cell. Therefore, the concentration of ^3He in the cell was constant, but the pressure changed with the temperature. The cell was installed in a low-temperature apparatus described elsewhere.[7] The temperature T of the top plate and the temperature difference ΔT between the top and bottom plates were measured with germanium thermometers. For the heat conduction measurements, T was regulated to $\pm 0.5\ \mu$K. The bottom was heated with a constant power.

Figure 1 shows the region of the stability diagram of the system[1] which was explored by our measurements. Changes in the reduced Rayleigh number $R/R_c(\psi = 0)$ correspond to changes in ΔT, whereas changes in the separation ratio ψ are accomplished by changes in the operating temperature T. The solid line labeled R_{co} is the Hopf bifurcation line, and that labeled R_{cs} corresponds to the stationary bifurcation. The two lines meet at the CT point.

Figure 2 shows the heat conduction of the cell as a function of ΔT for the three temperatures corresponding to the vertical dashed lines in Fig. 1. At the highest T, $\psi \cong 0.03$ and theory[1] indicates that the bifurcation to the convecting state should have occurred at $\Delta T \cong 3$ mK. However, that bifurcation is not visible because the initial slope of the heat conduction curve beyond it is smaller than that for a single-

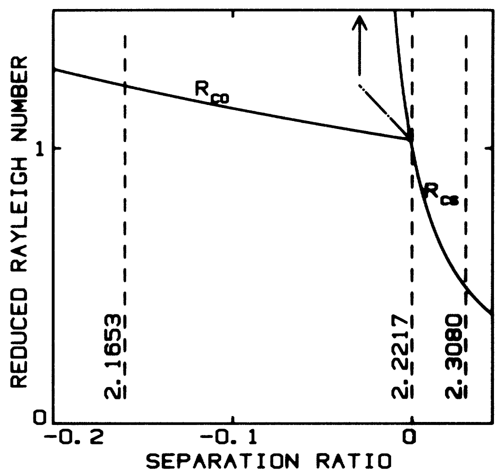

FIG. 1. Stability diagram of a mixture with $L=0.03$ contained in a porous medium and heated from below. The vertical dashed lines correspond approximately (see Ref. 6) to those experimental paths for which data are given in Fig. 2. The solid line labeled R_{co} is the Hopf bifurcation, and that labeled R_{cs} is the stationary bifurcation. At the position of the arrow, R_{cs} diverges. The dash-dotted line indicates the upper limit for the existence of the oscillations central to this paper.

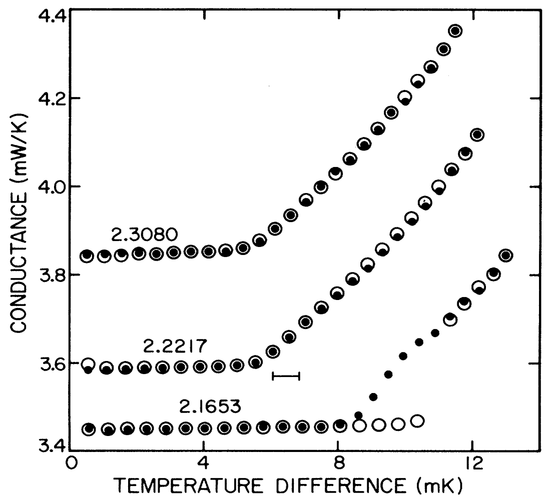

FIG. 2. The conductance of the cell as a function of the temperature difference ΔT between top and bottom plate when heated from below. The numbers given in the figure are the temperature of the top plate in kelvins. Open (solid) circles are taken with increasing (decreasing) ΔT. The small horizontal bar indicates the range of the oscillations shown in Fig. 4 below. It emphasizes that this range is small on the scale of Fig. 2.

component fluid by a factor of about $L^3/\psi \approx 10^{-3}$.[8] At ΔT near 6 mK, where the fluid would have undergone a bifurcation in the absence of the thermodiffusion effect ($\psi=0$), convection becomes apparent in the data. The heat conduction curve for $\psi=0.03$ shows no hysteresis and ΔT shows no oscillations, as expected for $\psi>0$.

At the lowest T (small expansion coefficient, $\psi \cong -0.16$), the measurements show hysteresis. As ΔT is increased from zero to 10.5 mK, the conductivity is almost constant. At $\Delta T = 10.5$ mK this purely conducting state becomes unstable against a hysteretic transition. The flow reached on the upper branch (after transients) is either steady or oscillatory, depending on ΔT. These phenomena are qualitatively similar to those observed for bulk binary mixtures in other experiments,[9–11] where they have been attributed[12] to a subcritical Hopf bifurcation. The oscillations that exist on the upper branch are not the ones relevant to the CT theory of Brand, Hohenberg, and Steinberg,[4] since they bifurcate from a nonlinear state. We observed the hysteresis illustrated in Fig. 2 ($T=2.1653$ K) for $\psi \lesssim -0.03$ and $T \lesssim 2.188$ K, but not for larger ψ and T.

For the middle temperature, $T=2.2217$ K, corresponding to $\psi \cong 0$, no oscillations are observed. This temperature, however, is very close to the CT point. Further reduction of T to 2.2216 K leads to oscillations setting in at a 6.1-mK temperature difference. These are the important ones for our work. They are described in more detail in Figs. 3, 4, and 5.

Figure 3 shows oscillations measured at $T=2.2064$ K. For small ΔT (6.364 mK and below) no oscillations are observed. They set in via a forward Hopf bifurcation between 6.364 and 6.371 mK. Further increase of ΔT leads to an increase of the amplitude and a de-

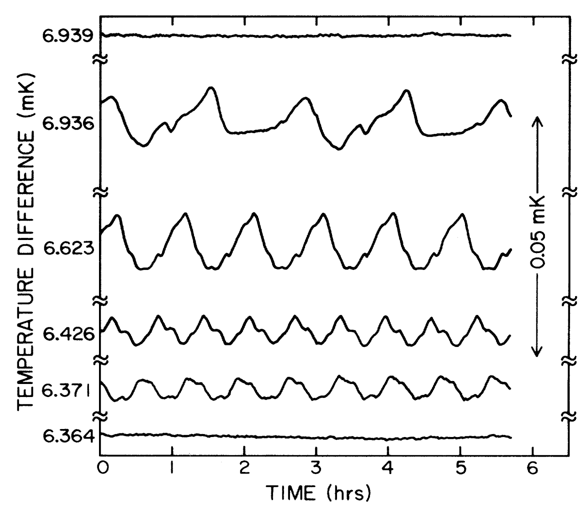

FIG. 3. The temperature difference across the cell, with the bottom plate heated at constant power, for six different values of that power. The numbers on the left indicate the mean temperature difference between top and bottom plate. The top temperature is 2.2064 K.

crease of the frequency. The smallest frequency is observed at a temperature difference of 6.936 mK. Although the time series shown for that ΔT covers only 2 cycles, corresponding to 6 h, we made measurements under similar conditions for 22 h and on the basis of those believe that the state is periodic. An increase by 3 μK of ΔT then leads to a steady flow state. This second bifurcation is accompanied by hysteresis: The temperature difference has to be lowered down to 6.84 mK to get the oscillatory state back. The approximate location of this hysteretic bifurcation is indicated in Fig. 1 by the dash-dotted line.

Figure 4 shows the frequency of the observed temperature oscillations as a function of ΔT for three different temperatures. The upper curve corresponds to Fig. 3. At larger T (2.2168 K), the range of oscillations is smaller and the frequencies are lower. This tendency continues with further increase of T (2.2197 K). For $T=2.2217$ K (see Fig. 2), no oscillations are observed because ψ has been raised beyond its CT value.

Figure 5 shows the frequency of the oscillations measured at the smallest temperature difference at which they exist, for different top temperatures. There are obviously two different branches. The fast oscillations (below 2.204 K) correspond to the case illustrated in Fig. 2 for 2.1653 K. They are reached via a hysteretic bifurcation. The interesting oscillations for this work are those of the slow mode observed at higher temperatures. Their frequency should correspond to the frequency given by linear stability analysis. It goes to zero at a temperature close to 2.22 K.

Close examination of the data in Fig. 2 at 2.2217 K, and similar measurements using pure ^{4}He, indicate that the bifurcation to the steady convective state is imperfect[13] in our cell (presumably because of horizontal temperature gradients caused by the porous medium, especially in the neighborhood of the lateral cell boundaries). This raises the question whether the main features of a codimension-two bifurcation exist in the presence of an imperfect bifurcation to the steady state. To shed some light on this problem we examined the equation

$$\ddot{W}-\alpha\dot{W}-\beta W+f_2W^2\dot{W}-f_1W^3+f_3W^5-h=0, \qquad (2)$$

which gives the amplitude W of the convective flow in the vicinity of the CT point.[4,14] Here the dots indicate time derivatives, and α and β are proportional to the vertical distances in Fig. 1 from the curves labeled R_{co} and R_{cs}, respectively. The fifth order term is added to stabilize[14] the solution for $\beta>0$ and the field h included to account for the "rounding"[13] in the experiment. Steady solutions W_0 of Eq. (2) are determined by

$$f_3W_0^5-f_1W_0^3-\beta W_0-h=0. \qquad (3)$$

Near the CT point, $f_1>0$ and thus for $h=0$, Eq. (3) corresponds to a backward bifurcation at $\beta=0$ (i.e., at R_{cs}).[4] In that case, there exist five real roots of Eq. (3) over the range $-f_1^2/4f_3<\beta<0$, corresponding to three stable and two unstable fixed points. For $|h|>0$, the bifurcation will be imperfect.[13] A linear stability analysis of the three stable fixed points, using Eqs. (2) and (3), shows that W_0 becomes unstable against small oscillations at

$$\alpha_c=f_2W_0^2, \qquad (4)$$

and that the frequency at onset is given by

$$\omega_c^2=-\beta-3f_1W_0^2+5f_3W_0^4=W_0(d\beta/dW_0). \qquad (5)$$

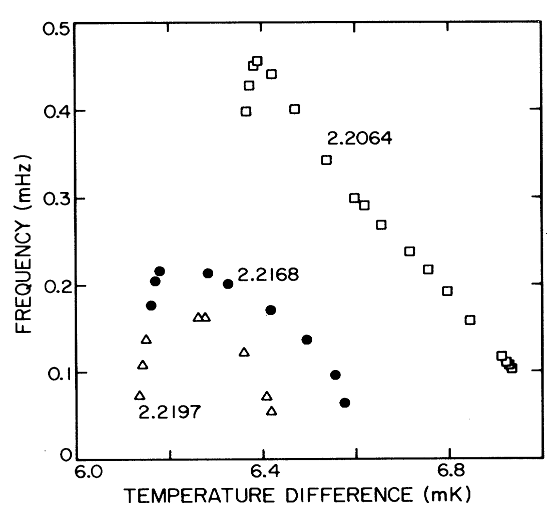

FIG. 4. The frequency of the temperature oscillations of the bottom plate as a function of the mean temperature difference for three different top temperatures. The labels indicate the temperature of the top in kelvins.

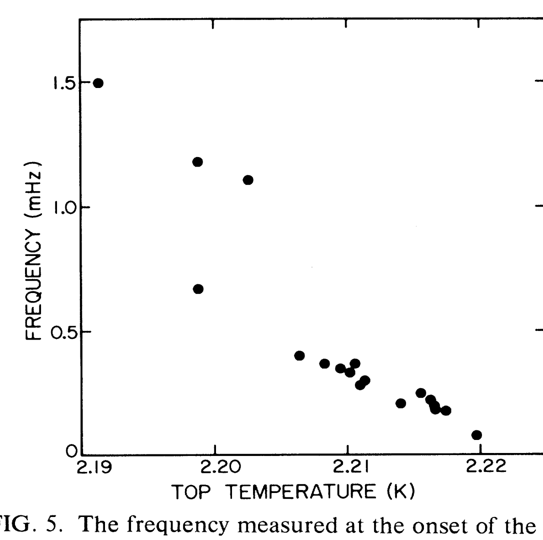

FIG. 5. The frequency measured at the onset of the oscillations as a function of the top temperature.

Thus, ω_c will vanish even for $|h|>0$ provided the curve $W_0(\beta)$ is folded so that $d\beta/dW_0$ vanishes. If, however, the field is large enough to destroy the hysteretic fold of that curve, the frequency of marginal oscillations will remain finite. In the experiment, ω_c goes to zero (Fig. 5), indicating that $|h|$ is sufficiently small for the hysteresis to survive. This also implies that the unstable fixed points corresponding to two of the roots of Eq. (3) still exist, and thus the orbit of the limit cycle generated at the Hopf bifurcation can expand with increasing α and pass through one of them, thereby yielding a vanishing frequency at finite amplitude as shown in Figs. 3 and 4.

Our evidence for a subcritical steady bifurcation in the neighborhood of the CT point is mostly indirect. The measurements support the existence of the unstable fixed point as discussed above. However, a direct measurement of the hysteresis loop at a top temperature of 2.2218 K (very close to the CT point) showed that its range was only approximately 10 μK. The difference in the conductance of the two stable branches was 1 μW/K or 0.03%. A bulk mixture (rather than a porous medium) will be preferable for observing the hysteresis. The smaller field will increase the range, and the absence of heat conduction through the nylon spheres will increase the relative difference of the cell conductance for the two branches.

An unresolved problem arising from this work is the fact that the frequency of the oscillations increases with ΔT in the neighborhood of the Hopf bifurcation (see Fig. 4). This is not caused by our constant–heat-flow measurement. We also performed the experiment by regulating the bottom temperature and measuring the heat current, and obtained the same oscillation frequency. Numerical integration of Eq. (2) with and without a field h did not show this behavior. Finally, we integrated the partial differential equation which includes the slow spatial variation of the order parameter [Eq. (3.9) of Ref. 4] and found that the frequency of the oscillations in the numerical experiment went down monotonically beyond the Hopf bifurcation, and neither vanishing nor the finite boundary conditions used by us led to an increase of the frequency similar to the one observed in the experiment.

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