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Codimension-2 bifurcations for convection in binary fluid mixtures

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An amplitude equation is derived for thermal convection in a binary fluid mixture in a porous medium and in bulk, in the vicinity of the intersection point of the lines of stationary and oscillatory instabilities. Slow spatial modulations are included in the amplitude equation near this codimension-2 bifurcation. The experimental realizability in binary fluid mixtures is discussed with use of data for specific systems such as alcohol-water mixtures and normal-fluid ^{3}He-^{4}He mixtures. Analogies are drawn to other physical systems which are easily accessible to experiment, such as the convective instability in nematic liquid crystals in an external magnetic field.

I. INTRODUCTION

Over the past few years the investigation of bifurcation phenomena in various branches of nonequilibrium physics, such as hydrodynamic instabilities, nonlinear quantum optics, autocatalytic chemical reactions, and population dynamics, has led to considerable progress in our understanding of these areas. The phenomenon of "codimension-2 bifurcations" has been investigated both from a mathematical viewpoint[1-6] and on the basis of simplified models of physical systems.[6-9] A codimension-2 bifurcation[5] occurs at a point in parameter space where two bifurcation lines intersect, thus leading to a competition between two types of instabilities. These instabilities might be oscillatory and stationary, but they might also both be oscillatory or stationary. From the mathematical investigations of this phenomenon it is known that different types of dynamic behavior can occur in the immediate vicinity of a single point in parameter space. Most of the models which have been analyzed so far have been highly idealized and thus not easily accessible to experiment. An example is the so-called thermohaline problem[3,6,7] (with or without an external magnetic field), in which a layer of fluid is subjected to vertical temperature and salinity gradients. The analysis assumes that one may vary concentration and temperature gradients *independently*, a condition which is difficult to meet in practice due to the cross coupling (thermodiffusion) between the gradients which exist in real fluids.

The goal of this paper is to give a detailed derivation of the amplitude equation near the intersection point for the stationary and oscillatory instabilities which occur in a layer of a binary fluid mixture with and without a porous medium and subject to a vertical temperature gradient. In a previous paper[9] we have given this amplitude equation without any derivation. The advantage of the system investigated here is that it is both experimentally realizable and amenable to a simple theoretical treatment. Indeed, in contrast to the thermohaline system, one has two independently variable parameters, the applied temperature gradient (or Rayleigh number) and the separation ratio ψ. This parameter contains the dissipative cross coupling between concentration and temperature gradients, the thermal-expansion coefficient, and the derivative of the density with respect to the concentration. The separation ratio can assume either sign and it can be changed experimentally, for instance by varying the relative concentrations of the components of the mixture. Emphasis will also be laid on the discussion of suitable boundary conditions and on the presentation of data for some relevant experimental systems.

For the first time in the study of codimension-2 bifurcation problems we will include in our description slow spatial modulations of the envelope function, albeit in one dimension only, thus generalizing the equations of Segel,[10] and of Newell and Whitehead[10] (which were derived for the onset of convection in a simple fluid). The new feature which arises is a term containing a first derivative in time and a second derivative in space, i.e., $\partial_t \partial_x^2 W$, where W is the envelope function.

Having treated convection in binary mixtures[11,12] we also discuss briefly convective instabilities in nematic liquid crystals.[13] This system is more complicated than a binary fluid mixture in that the structure of the basic equations is anisotropic in space and is characterized by nonlinearities of infinite order. Nematic liquid crystals are attractive experimentally, however, since one can use as a second continuously variable parameter a static magnetic field which changes the elastic energy and thus the frequency and the Rayleigh number. Indeed, an experiment has been performed recently[14] using precisely this technique. The major problem with interpreting the data is the fact that the structure which occurs above the threshold for the onset of the oscillatory instability is as yet not well characterized.

This paper is organized as follows. In Sec. II we derive in detail the amplitude equation without spatial modulation for a porous system and for bulk binary mixtures with stress-free horizontal boundaries. We then study the influence of various types of boundary conditions on these systems and describe the dynamical behavior expected near the codimension-2 bifurcation point. In Sec. III the influence of slow spatial modulations is investigated, and some implications are discussed. In Sec. IV we present quantitative estimates for real binary mixtures at room temperature and low temperatures, as well as a discussion of other experimentally relevant systems such as nematic or cholesteric liquid crystals. A number of detailed calculations are given in the Appendices.

II. THE AMPLITUDE EQUATION WITHOUT SLOW SPATIAL MODULATIONS

In this section we start from the nonlinear equations for the deviations from the heat-conduction state and derive the amplitude equation near the polycritical surface. In Sec. II A we discuss the derivation for a mixture in a porous medium and in Sec. II B we consider bulk binary mixtures. We use two different methods to obtain the amplitude equation. The first approach makes use of the amplitude equations derived previously[11] for the stationary and the oscillatory instabilities, respectively. Assuming the structure of the resulting amplitude equation, it becomes possible by taking the appropriate limits to evaluate its coefficients. The second technique, which is more satisfactory from a logical point of view since it does not assume the form of the answer, starts off from the basic nonlinear equations[12] and carries out a projection onto the generalized eigenspace of the linearized problem near the codimension-2 surface. The first approach is simpler and more intuitive. The second is quite general and provides a sound basis for the first method. For mixtures in a porous medium we give all details of the derivation (with both approaches) in Appendix A. For bulk mixtures, which are discussed in Sec. II B and in Appendix B, we outline the derivation using the second approach. In Sec. II C the temporal behavior of the dynamical system near the bifurcation point is discussed, and in Appendix D the nonlinear oscillatory solutions of the amplitude equations are calculated. Finally, in Sec. II D we consider the changes which can be expected in the results if realistic boundary conditions are imposed,[12,15–20] rather than the somewhat idealized ones we have assumed, especially for the case of bulk binary mixtures.

A. Binary mixtures in a porous medium

The basic nonlinear equations for the deviations from the heat-conduction state of a binary mixture of miscible fluids in a porous medium read, in dimensionless units,[12]

$$[(K\kappa/l^2\epsilon\nu)\partial_t+1]\Delta w-\Delta_2\theta-\Psi\Delta_2 c=0\ , \tag{2.1}$$

$$Rw+(-\partial_t+\Delta-\vec{v}\cdot\vec{\nabla})\theta=0\ , \tag{2.2}$$

$$Rw+[(D/\kappa)\Delta-\partial_t-\vec{v}\cdot\vec{\nabla}]c-(D/\kappa)\Delta\theta=0\ , \tag{2.3}$$

where w is the z component of the velocity field $\vec{v}$, θ is the deviation of the temperature from the conduction profile, c is the concentration of one component, R is the Rayleigh number $R=\beta_1 glK\,\Delta T/\nu\kappa$ corresponding to the temperature difference ΔT, $\Psi=-k_T\beta_2/T\beta_1$ is the separation ratio, $\beta_1=-\rho^{-1}(\partial\rho/\partial T)_{P,c}$ is the thermal-expansion coefficient, $\beta_2=-\rho^{-1}(\partial\rho/\partial c)_{P,T}$, κ and D are the thermodiffusivity and the diffusion coefficient, k_T is the thermodiffusion ratio,[21] $\Delta_2=\partial^2/\partial x^2+\partial^2/\partial y^2$ is the horizontal part of the Laplace operator Δ, K is the permeability, ϵ is the porosity, ν is the kinematic viscosity, and l is the height of the layer. Time is scaled with l^2/κ, velocity with κ/l, temperature with $\nu\kappa/\beta_1 glK$, and concentration with $-\nu\kappa k_T/T\beta_1 gKl$, and we shall choose units of length such that $l=1$. Equations (2.2) and (2.3) are identical to the corresponding equations in binary fluid mixtures (in dimensionless units). Equation (2.1) replaces the equation for the velocity field and the incompressibility condition for usual binary mixtures. In deriving this equation one makes use of Darcy's law for the averaged velocity field.[22] Contrary to usual hydrodynamics, the velocity field in a porous medium is not a Galilean invariant quantity and thus it is impossible to prescribe the gradients of the velocity field on the boundaries ($\vec{v}$ satisfies only a first-order differential equation with respect to the spatial coordinates).

The boundary conditions we take in this section for the velocity field $\vec{v}=(u,v,w)$ are

$$\begin{aligned} &u=0 \quad \text{at } x=0 \text{ and } x=L_x\ ,\\ &v=0 \quad \text{at } y=0 \text{ and } y=L_y\ ,\\ &w=0 \quad \text{at } z=0 \text{ and } z=1\ , \end{aligned} \tag{2.4}$$

where we have assumed a rectangular container with sides L_x, L_y, and L_z $(=l=1)$. Different boundary conditions for the velocity field will be discussed in Sec. II D. For temperature and concentration fluctuations we assume

$$\begin{aligned} &\partial_x\theta=\partial_x c=0 \quad \text{at } x=0 \text{ and } x=L_x\ ,\\ &\partial_y\theta=\partial_y c=0 \quad \text{at } y=0, \text{ and } y=L_y\ ,\\ &\theta=c=0 \quad \text{at } z=0 \text{ and } z=1\ . \end{aligned} \tag{2.5}$$

The boundary conditions for the concentration field are an idealization of the more physical "no-mass-flux" boundary condition.[12,15–20] As will be discussed in Sec. II D, however, none of the features presented in this section are expected to change qualitatively when the more realistic boundary conditions are used.

To arrive at the results for linear stability we proceed as usual.[12] Inserting the ansatz[9]

$$\begin{bmatrix} w(x,z,t) \\ \theta(x,z,t) \\ c(x,z,t) \end{bmatrix} = \begin{bmatrix} W(t) \\ \Theta(t) \\ C(t) \end{bmatrix} \sin(\pi z)\cos(\pi x)\ , \tag{2.6}$$

we obtain, after setting $W(t)=w_1\exp(\sigma t)$, a cubic polynomial for σ of the form

$$a_3\sigma^3+a_2\sigma^2+a_1\sigma+a_0=0\ . \tag{2.7}$$

For the stationary instability ($\sigma\equiv 0$) we have the critical Rayleigh number[12]

$$R_{cs}=4\pi^2[1+\psi(1+L^{-1})]^{-1}\ , \tag{2.8}$$

where we have introduced the Lewis number

$$L\equiv D/\kappa\ , \tag{2.9}$$

and the critical-wave number is $k_{c\perp}=\pi$. For the oscillatory instability we find a critical Rayleigh number[12]

$$R_{co}=4\pi^2(1+L)(1+\psi)^{-1} \tag{2.10a}$$

and a frequency at onset

$$\omega_o^2=-4\pi^4L^2[1+\psi(1+L^{-1}+L^{-2})](1+\psi)^{-1}\ . \tag{2.10b}$$

The polycritical point is defined by $R_{\rm pc}=R_{co}=R_{cs}$, which yields

$$R_{\rm pc}=4\pi^2(1+L+L^2)\ , \tag{2.11a}$$

$$\omega_{\rm pc}=0\ , \tag{2.11b}$$

$$\psi_{\rm pc}=-L^2(1+L+L^2)^{-1}\ . \tag{2.11c}$$

As is obvious from Eq. (2.11c) the separation ratio for which the crossover between stationary and oscillatory instability occurs depends sensitively on the thermal diffusivity and the diffusion coefficient. For water-alcohol mixtures one has typically $\kappa/D=L^{-1}\approx 100$, which means that $\psi_{\rm pc}\approx -10^{-4}$, whereas for ^{3}He-^{4}He mixtures $L^{-1}\approx 2$ (near the tricritical point) and one can get $\psi_{\rm pc}\simeq -0.15$. It is well established[16,12] that the first instability to occur is stationary for $\psi>\psi_{\rm pc}$ and oscillatory for $\psi<\psi_{\rm pc}$, with vanishing oscillation frequency $\omega_{\rm pc}$ and $\psi_{\rm pc}$. The third root of Eq. (2.7) is negative so that it is possible to neglect the term $\sim\sigma^3$ for sufficiently small σ. Thus one expects near the polycritical surface an equation of the form[23]

$$\ddot{W}-\alpha\dot{W}-\beta W+f(W,\dot{W})=0\ , \tag{2.12}$$

where f is the nonlinear function of W and $\dot{W}$. Due to the up-down symmetry of the present problem, f cannot contain terms such as W^2, $\dot{W}^2$, or $W\dot{W}$. Thus one would expect that to lowest order in the nonlinearity, f will have terms of the form W^3, $W^2\dot{W}$, $\dot{W}^2W$, and $\dot{W}^3$. We show in Appendix C that only the terms $W^2\dot{W}$ and W^3 are necessary. We shall therefore seek the prefactors of those to complete the determination of the amplitude equation, which reads

$$\ddot{W}-\alpha\dot{W}-\beta W+f_2W^2\dot{W}-f_1W^3=0\ , \tag{2.13}$$

with

$$\alpha=2\pi^2(1+L)(R-R_{co})/R_{co} \tag{2.14}$$

and

$$\beta=4\pi^4L(R-R_{cs})/R_{cs}\ , \tag{2.15}$$

coming from the linear analysis. In Eqs. (A12), (A19), (A43), and (A49) of Appendix A we derive the following expressions for the coefficients:

$$f_1=\pi^2/4\ , \tag{2.16}$$

$$f_2=\tfrac{1}{4}(L^{-1}+1)\ . \tag{2.17}$$

Equation (2.13) with Eqs. (2.14)–(2.17), constitute the main result of this section. Its physical implications are discussed in Sec. II C and Appendix D. A simple general argument to determine the minimal form of the amplitude equation (2.13) is given in Appendix C, for both codimension-2 and codimension-3 bifurcations.

B. Bulk binary fluid mixtures

For bulk mixtures we introduce the velocity potential $\bar\psi$ with $w=\partial\bar\psi/\partial x$ and $u=-\partial\bar\psi/\partial z$ and then have for the basic equations[15–20]

$$\frac{1}{P}\left[\frac{\partial\bar\psi}{\partial x}\frac{\partial\Delta\bar\psi}{\partial z}-\frac{\partial\bar\psi}{\partial z}\frac{\partial\Delta\bar\psi}{\partial x}\right]+\Delta\left[\frac{1}{P}\frac{\partial}{\partial t}-\Delta\right]\bar\psi=\Delta_2\theta+\psi\Delta_2c\ , \tag{2.18a}$$

$$R\frac{\partial\bar\psi}{\partial x}-L\Delta\theta+\left[L\Delta-\frac{\partial}{\partial t}\right]c-\frac{\partial\bar\psi}{\partial x}\frac{\partial c}{\partial z}+\frac{\partial\bar\psi}{\partial z}\frac{\partial c}{\partial x}=0 \tag{2.18b}$$

$$R\frac{\partial\bar\psi}{\partial x}+\left[-\frac{\partial}{\partial t}+\Delta\right]\theta-\frac{\partial\bar\psi}{\partial x}\frac{\partial\theta}{\partial z}+\frac{\partial\bar\psi}{\partial z}\frac{\partial\theta}{\partial x}=0\ . \tag{2.18c}$$

In Eqs. (2.18) we have used the same notation as in Eqs. (2.1)–(2.3), but we have introduced a new parameter, the Prandtl number,

$$P\equiv\nu/\kappa\ , \tag{2.19}$$

which involves the kinematic viscosity ν. The ratio of Prandtl and Lewis numbers is known as the Schmidt number,

$$S\equiv P/L=\nu/D\ . \tag{2.20}$$

Taking into account the relationship between $\bar\psi$ and u,w it is immediate that Eqs. (18b) and (18c) and (2.2) and (2.3) coincide, whereas the dynamic equation for the velocity (2.18a), which already incorporates the incompressibility condition, is markedly different from Eq. (2.1). In the case of bulk mixtures we consider a fluid layer of infinite lateral extent. At the top and bottom plates we assume free-slip boundary conditions for the vertical velocity field w,

$$w=\frac{d^2w}{dz^2}=0,\quad \text{for } z=0,1 \tag{2.21}$$

and for concentration and temperature fluctuations we take

$$\theta=c=0,\quad \text{for } z=0,1\ . \tag{2.22}$$

In Sec. II D we shall discuss the effect of more realistic boundary conditions (rigid, no-mass-flux). Adopting the standard procedure for linearized stability analysis we obtain a polynomial for the growth rate σ of the form (2.7).

At the stationary instability ($\sigma \equiv 0$) we have for the critical Rayleigh number R_{cs}

$$R_{cs} = [(k_{\parallel}^2 + k_{\perp c}^2)^3 / k_{\perp c}^2][1 + \psi(1+L)^{-1})]^{-1} , \qquad (2.23)$$

with $k_{\parallel} = \pi$ and $k_{\perp c} = \pi/\sqrt{2}$. For the oscillatory instability [$\mathrm{Re}\sigma = 0$, $\mathrm{Im}\sigma \neq 0$] we have

$$R_{co} = \frac{(k_{\parallel}^2 + k_{\perp c}^2)^3}{k_{\perp c}^2} \frac{(1+L)(1+L+P+LP^{-1})}{1+P(1+\psi)} , \qquad (2.24a)$$

and

$$\omega_o = (k_{\parallel}^2 + k_{\perp c}^2) L \{ -\psi(1+L^{-1})(1+PL^{-1}) \times [1+P(1+\psi)]^{-1} - 1 \}^{1/2} . \qquad (2.24b)$$

For the intersection of the lines of stationary and oscillatory instabilities, i.e., for the polycritical point, we find $R_{co} = R_{cs} = R_{\mathrm{pc}}$, with

$$R_{\mathrm{pc}} = [(k_{\parallel}^2 + k_{\perp c}^2)^3 / k_{\perp c}^2][1 + (1+L^{-1})\psi_{\mathrm{pc}}]^{-1} , \qquad (2.25)$$

$$\psi_{\mathrm{pc}} = -(1+P)[1+P+L^{-1}+P/L+P/L^2]^{-1} , \qquad (2.26)$$

$$\omega_{\mathrm{pc}} = 0 . \qquad (2.27)$$

Proceeding as in the case of a binary fluid mixture in a porous medium, one is led to an equation near the polycritical point of the form

$$\ddot{W} - \alpha' \dot{W} - \beta' W + f(W, \dot{W}) = 0 , \qquad (2.28)$$

where $f(W, \dot{W})$ is the same nonlinear function as in Sec. II A,

$$f(W, \dot{W}) = f_2' W^2 \dot{W} - f_1' W^3 , \qquad (2.29)$$

i.e., Eqs. (2.28) and (2.13) are isomorphic, only the numerical values of the coefficients are different. For f_1' and f_2' we find in bulk binary fluid mixtures [cf. Eqs. (B11) and (B12) of Appendix B]

$$f_1' = \tfrac{3}{32}\pi^4 , \qquad (2.30)$$

$$f_2' = \tfrac{7}{64}\pi^2[1+(1+P)(L^2+L+PL)^{-1}] . \qquad (2.31)$$

From Eq. (2.31) we see that in the limit of infinite kinematic viscosity the expression becomes proportional to $(1+L^{-1})$, i.e., the dependence on $L = D/\kappa$ is the same as in the porous medium case. This is not surprising because a fluid mixture in a porous medium represents a highly confined situation which can roughly be thought of as a very viscous fluid. Since both f_1' and f_2' have the same sign as in the porous medium case, the essential difference is that the location of the separation line between the sectors IIa and IIb of Fig. 1 now depends on two parameters, the Lewis number $L = D/\kappa$ (as in porous media) as well as the Prandtl number P.

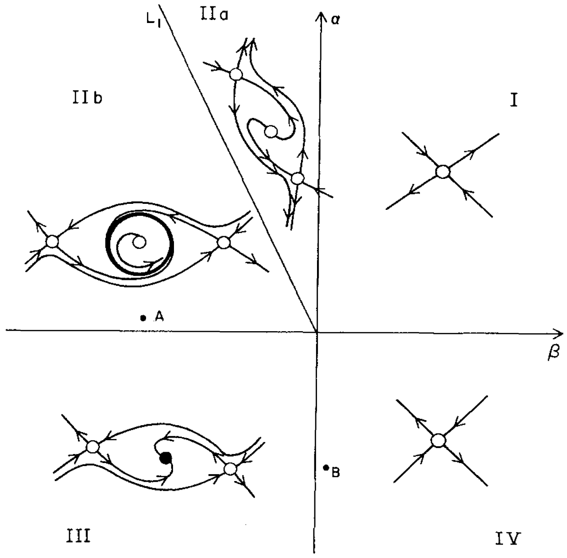

FIG. 1. Schematic phase portrait of the behavior of Eq. (2.13) in the vicinity of the polycritical point $\alpha = \beta = 0$. The parameter space is divided into sectors with different characteristic behavior. In each sector we show typical phase-space orbits and the stability of various attractors (the stable fixed point is shown as a solid circle, the unstable ones are shown as open circles, and the stable limit cycle is a thick solid line; orbits are drawn as thin solid lines). Quadrant III has one stable and two unstable fixed points, while quadrants I and IV have one unstable fixed point, with an inverted stationary bifurcation along the line $\beta = 0$, $\alpha < 0$. On the line $\alpha = 0$, $\beta < 0$ the system makes a forward oscillatory bifurcation to a stable limit cycle, which disappears along the line L_1 where the oscillation period diverges. In sector IIa there are three unstable points joined by a heteroclinic orbit. The points A and B are discussed in Appendix A 1.

In closing this subsection we note that the nonlinear coefficients [(2.30) and (2.31)] imply a Nusselt number N which is different from the one obtained by Gutkowicz-Krusin, Collins, and Ross.[18,19] Indeed, the general expression[24] for N in terms of the velocity and temperature may be evaluated immediately above the threshold for the stationary instability using the formulas of Appendix B valid for stress-free horizontal boundaries. The result we find is

$$N - 1 = 2[(R - R_{cs})/R_{cs}][1 + \psi(1+L^{-2})] \times [1+\psi(1+L^{-2}+L^{-3})]^{-1} . \qquad (2.32)$$

This formula should hold wherever the stationary instability leads to a forward (not inverted) bifurcation, a condition which is, however, not realized near the polycritical point. The Nusselt number obtained from Eq. (65) of Gutkowicz-Krusin *et al.*,[18] for stress-free boundaries, is given by a different formula:

$$N - 1 = 2[(R - R_{cs})/R_{cs}](1+\psi)(1+3\psi L^{-2})^{-1} . \qquad (2.33)$$

The error in the above result comes from the assumption, originally made by Chandrasekhar[25] that the nonlinear behavior above threshold can be obtained purely from the linear modes. It is well known, however, even for the case of convection in simple fluids,[24] that in lowest order the modification of the vertical profile may not be neglected in calculating the Nusselt number.

C. Dynamical behavior resulting from the amplitude equation

In this subsection we review the dynamical behavior predicted by the amplitude equation (2.13) in the vicinity of the codimension-2 point $\alpha=\beta=0$, for fixed f_i (which are positive and of order unity). The general behavior of equations having double zero eigenvalues was first elucidated by Takens[1] and Arnold[2] and has been the subject of numerous investigations by mathematicians (see Refs. 3, 5, and 6 for reviews). The steady solutions of Eq. (2.13) are fixed points at $W=0$, $\dot{W}=0$ and at $W=\pm(-f_1/\beta)^{1/2}$, $\dot{W}=0$, and a periodic solution (limit cycle) which only occurs for $\alpha>0$, $\beta<0$ (see Fig. 1). In order to determine the stability of these solutions we must linearize the equations about each solution and evaluate the dynamical eigenvalues. The procedure is simplest for the $W=\dot{W}=0$ fixed point whose eigenvalues are

$$\sigma_\pm=\tfrac{1}{2}[\alpha\pm(\alpha^2+4\beta)^{1/2}] , \tag{2.34}$$

from which it follows that this fixed point is only stable ($\mathrm{Re}\sigma_\pm<0$) in the third quadrant ($\alpha<0,\beta<0$). The stability of the fixed points at $W=\pm(-f_1/\beta)^{1/2}$ can be studied by a similar linearization. The result, indicated in Fig. 1, is that these fixed points are always unstable for $f_1>0$. On crossing the $\alpha=0$ axis upward for $\beta<0$ into the second quadrant, Eq. (2.34) shows that the $W=0$ fixed point has an oscillatory instability [$\mathrm{Im}\sigma_\pm\neq 0$], and it can be shown that Eq. (2.13) has a limit-cycle solution, at least sufficiently close to the horizontal axis. The domain of existence of this solution can be studied analytically for small α and β. As shown in Appendix D the limit cycle disappears when the orbit $W(t)$ grows sufficiently large to pass through the fixed points at $W=\pm(-f_1/\beta)^{1/2}$. This occurs along the line L_1 of Fig. 1, given by

$$\alpha=-(f_2/5f_1)\beta . \tag{2.35}$$

The analysis of Apendix D may also be extended to show that the limit-cycle solution is stable in its domain of existence.

The full dynamical behavior around the polycritical point is shown in Fig. 1, where the various solutions and their stability are indicated, as well as the transitions (bifurcations) between solutions as one changes α and β. Because of the signs of the coefficients f_1 and f_2, only the sectors III and IIb have stable solutions. In the sectors IIa, I, and IV, Eq. (2.13) has no stable solutions and $W(t)$ and $\dot{W}(t)$ will in general grow, thus invalidating the perturbation theory which led to (2.13). In order to find the precise state of the system in those sectors it is necessary to solve the full system [(2.1)–(2.3)], and the proximity to the polycritical point does not *a priori* simplify the analysis. We are therefore able to predict a Hopf bifurcation along the negative β axis and an inverted bifurcation in crossing the negative α axis from region III to region IV where the precise behavior is unknown. Along the line L_1 the limit cycle slows down to zero frequency, with a bifurcation to some other behavior [not determined by Eq. (2.13)] in sector IIa. Note that throughout sector IIa there exists a heteroclinic orbit[5] joining the three unstable fixed points in the system. The appearance of such orbits is often the precursor to a transition to chaos. The multiplicity of behavior in the immediate vicinity of a single point in parameter space should be experimentally observable if α and β can be sufficiently well controlled. Physically, this means controlling the temperature difference which determines R and the mean concentrations of the components, which control ψ.

D. Realistic boundary conditions

Up to now we have assumed that the concentration is kept constant at the upper and lower boundaries, Eqs. (2.5) and (2.22). The question naturally arises to what extent the situation outlined above carries over if more realistic boundary conditions for the concentration field are implemented (no-flux boundary condition). Let us first consider bulk binary mixtures. It has been shown by many authors[15–17,20] that different boundary conditions in this case do not change the qualitative picture, i.e., the point of intersection of stationary and oscillatory branches exists for any boundary conditions.

If we introduce the variable

$$\eta=c-\theta , \tag{2.36}$$

we can obtain from the relation

$$J^c=\nabla c-\nabla\theta=0 \tag{2.37}$$

(in dimensionless units) the no-flux boundary condition

$$\frac{d\eta}{dz}=0, \quad \text{at } z=0 \text{ and } z=1 . \tag{2.38}$$

Taking into account the rigid boundary conditions for the velocity field

$$w=\frac{dw}{dz}=0, \quad \text{at } z=0 \text{ and } z=1 \tag{2.39}$$

together with (2.38) we obtain a linearized boundary value problem very similar to that treated in Ref. 16,

$$\left[\frac{d^2}{dz^2}-k_\perp^2\right]\left[\frac{\sigma}{P}-\frac{d^2}{dz^2}+k_\perp^2\right]w=-k_\perp^2[\theta(1+\psi)+\psi\eta] , \tag{2.40}$$

$$\left[\sigma-\frac{d^2}{dz^2}+k_\perp^2\right]\theta=Rw , \tag{2.41}$$

$$\left[\sigma-L\left[\frac{d^2}{dz^2}-k_\perp^2\right]\right]\eta=-\left[\frac{d^2}{dz^2}-k_\perp^2\right]\theta , \tag{2.42}$$

where we have introduced

$$(w,\theta,\eta)=(w(z),\theta(z),\eta(z))e^{i(\vec{k}_\perp\cdot\vec{r})+\sigma t} . \tag{2.43}$$

Contrary to the boundary value problems studied in Secs. II A and II B above, the present one is nonseparable and cannot be solved analytically. Using a Galerkin technique analogous to one first used in Ref. 17, one arrives at an equation for σ of the form

$$a_3\sigma^3+a_2\sigma^2+a_1\sigma+a_0=0 , \tag{2.44}$$

with

$$a_3=-\frac{1}{P}\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]w\,dz\;, \tag{2.45a}$$

$$a_2=(1+P^{-1}+LP^{-1})\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]^2 w\,dz\;, \tag{2.45b}$$

$$a_1=-(1+\psi)Rk_\perp^2\int_0^1 w^2dz-(1+L+LP^{-1})\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]^3 w\,dz\;, \tag{2.45c}$$

$$a_0=L\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]^4 w\,dz+Rk_\perp^2\int_0^1\left[\frac{d^2}{dz^2}-k_\perp^2\right]w[\psi+(1+\psi)L]dz\;. \tag{2.45d}$$

One can use Eqs. (2.44) and (2.45) to obtain the critical values of R, $k_\perp$, and σ for any specific system by choosing trial functions which satisfy the appropriate boundary conditions. There are two ways to check the accuracy of the calculations. The first possibility is to calculate the trial function in higher approximations and to check the convergence of the Galerkin procedure. The second possibility is to compare the result with an exact solution, at least in some limit if it is available. In the case of the stationary instability the first method has been used in Ref. 17 and the second method in Ref. 16. However, in the case of the oscillatory instability the first method presents considerable complications, since no exact solution is known. Therefore, in order to check the accuracy and reliability of the procedure, two different trial functions were used in Ref. 17, and two different expressions for R_c and ω_c were used in Ref. 16. It should be noted that there is no variational principle for the oscillatory instability in binary mixtures, i.e., it is impossible to prove that R_c is the extremum of a functional, so no strict upper bound to the exact value is known (such results exist primarily for self-adjoint operators). What is possible is the calculation performed in Ref. 16, which shows that the results for R_c, k_c, and ω_c agree closely with each other, even in the first approximation, using two different trial functions in the Galerkin method. Also, one of the expressions reduces to the variational estimate for stationary convection in the limit $\omega=0$. As has been shown in Ref. 16, in the region of parameter space of interest here (the vicinity of the polycritical point), different boundary conditions do not change the picture qualitatively. The main effect is an overall shift of the values of R_c and $k_{\perp c}$. It is only far away from the codimension-2 bifurcation that the transverse wave number for the onset of the stationary instability changes drastically and is determined by the container size in the case of no-flux boundary conditions.[16]

The same argument can be given for the case of binary mixtures in porous media. Since the velocity and temperature boundary conditions in (2.4) and (2.5) are the physically correct ones, we need only implement the no-flux condition, i.e., replace the concentration boundary condition in (2.5) by (2.37). Then, using η defined in Eq. (2.36) as a variable (instead of c), we have for the linearized basic equations [(2.1)–(2.3)]

$$\frac{-\kappa K}{l^2\epsilon\nu}\sigma\left[\frac{d^2}{dz^2}-k_\perp^2\right]w =\left[\frac{d^2}{dz^2}-k_\perp^2\right]w+k_\perp^2[(1+\psi)\theta+\psi\eta]\;,$$

$$\left[\sigma-\frac{d^2}{dz^2}+k_\perp^2\right]\theta=Rw\;, \tag{2.46}$$

$$\left[\sigma-L\left[\frac{d^2}{dz^2}-k_\perp^2\right]\right]\eta=-\left[\frac{d^2}{dz^2}-k_\perp^2\right]\theta\;.$$

Proceeding as in the case of a bulk binary mixture we once again obtain a critical polynomial of the form (2.44), with coefficients

$$a_3=\frac{\kappa K}{l^2\epsilon\nu}\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]w\,dz\;, \tag{2.47a}$$

$$a_2=\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]w\,dz -\frac{\kappa K}{l^2\epsilon\nu}(1+L)\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]^2 w\,dz\;, \tag{2.47b}$$

$$a_1=\frac{K\kappa L}{l^2\epsilon\nu}\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]^3 w\,dz -(1+L)\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]^2 w\,dz +k_\perp^2(1+\psi)R\int_0^1 w^2dz\;, \tag{2.47c}$$

$$a_0=-k_\perp^2L(1+\psi+\psi L^{-1})R\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]w\,dz +L\int_0^1 w\left[\frac{d^2}{dz^2}-k_\perp^2\right]^3 w\,dz\;. \tag{2.47d}$$

As above one can use (2.47) to evaluate the critical values R_c, ω_c, and $k_{\perp c}$ (after R_c has been minimized also with respect to $k_\perp$) by choosing appropriate trial functions for any specific system under consideration. It should be stressed that the values one gets from carrying out this procedure are of relevance to real experiments because they correspond to realistic vertical *and* lateral boundary conditions. As above we believe that the results will be qualitatively the same as those derived analytically in Sec. II A.

III. AMPLITUDE EQUATION WITH SLOW SPATIAL MODULATIONS

For convection in layers of large horizontal extent the flow pattern is highly degenerate since only the magnitude of the transverse wave vector is fixed but not its direction. In such a situation an amplitude equation of the form (2.13), which involves only a single mode $W(t)$ with rolls parallel to the y direction, is clearly an oversimplification. Even assuming that the rolls stay parallel to y, it is possible to make small departures from the critical wave vector by a small distortion of the flow. For the case of convection in a simple fluid, Segel[10] and Newell and Whitehead[10] have shown how the one-mode amplitude equation is changed by spatial modulation. In the simple case of parallel rolls the complex amplitude $\overline{w}(t,x)$ is given by

$$\tau \frac{\partial \overline{w}}{\partial t} = \left[\frac{R - R_c}{R_c} \right] \overline{w} + \xi^2 \frac{\partial^2 \overline{w}}{\partial x^2} - g \mid \overline{w} \mid^2 \overline{w} \;, \tag{3.1}$$

where

$$\xi^2 = \frac{1}{2} \frac{\partial^2 R_c(k_\perp)}{\partial k_\perp^2} \Bigg|_{k_\perp = k_{\perp c}} \tag{3.2}$$

depends on the deviations of $R_c(k_\perp)$ from the critical value $R_c(k_{\perp c})$. We wish to generalize Eq. (3.1) to the case of codimension-2 bifurcations treated in Eq. (2.13). For simplicity we restrict ourselves to the case of parallel rolls (two-dimensional flow) where the amplitude function depends again on only one spatial coordinate.

Near the stationary bifurcation we have the generalization of Eq. (A1) of Appendix A,

$$\tau_s \dot{w}_s = \epsilon_s w_s + \xi_s^2 \frac{\partial^2 w_s}{\partial x^2} - g_s \mid w_s \mid^2 w_s \;, \tag{3.3}$$

with[26]

$$\xi_s^2 = \frac{1}{2} \frac{\partial^2 R_{cs}(k_\perp)}{\partial k_\perp^2} \Bigg|_{k_\perp - k_{\perp c}} . \tag{3.4}$$

Near the oscillatory bifurcation we have $\overline{w} = w_o$ with

$$W(t,x) = w_o(t,x) e^{i\omega_o t} + \text{c.c.} \;, \tag{3.5}$$

and the amplitude equation reads, instead of (A6),

$$\tau_o \frac{\partial w_o}{\partial t} = \epsilon_o w_o + \xi_o^2 \frac{\partial^2 w_o}{\partial x^2} - g_o \mid w_o \mid^2 w_o \;, \tag{3.6}$$

with

$$\xi_o^2 = \frac{1}{2} \frac{\partial^2 R_{co}(k_\perp)}{\partial k_\perp^2} \Bigg|_{k_\perp = k_{\perp c}} . \tag{3.7}$$

These generalizations of (A1) and (A6) can be obtained by making the replacements[26]

$$\epsilon_s \rightarrow \epsilon_s + \xi_s^2 \frac{\partial^2}{\partial x^2} \;, \tag{3.8a}$$

$$\epsilon_s \rightarrow \epsilon_o + \xi_o^2 \frac{\partial^2}{\partial x^2} \tag{3.8b}$$

in the appropriate amplitude equations (A1) and (A6). It is thus clear that to leading order in the spatial variations the amplitude equation (2.13) is replaced by

$$\ddot{W} - \left[\tilde{\alpha} \left[\epsilon_o + \epsilon_o^2 \frac{\partial^2}{\partial x^2} \right] - f_2 \mid W \mid^2 \right] \dot{W} - \left[\tilde{\beta} \left[\epsilon_s + \xi_s^2 \frac{\partial^2}{\partial x^2} \right] + f_1 \mid W \mid^2 \right] W = 0 \;, \tag{3.9}$$

where $\tilde{\alpha}\epsilon_0 = \alpha, \tilde{\beta}\epsilon_s = \beta$, and the function $W(x,t)$ is now, in general, complex and defined by

$$w(x,z,t) = \tfrac{1}{2} W(x,t) \sin(\pi z) \exp(i\pi x) + \text{c.c.} \;, \tag{3.10}$$

which replaces Eq. (2.6).

Equation (3.9) has a mathematical structure which may lead to interesting behavior, due especially to the presence of the mixed term $\partial^3 W / \partial t \partial x^2$. In particular, this lowest-order equation does not derive from any minimizing potential.

IV. QUANTITATIVE ESTIMATES AND POSSIBLE EXPERIMENTS

Since we are interested in the behavior of normal-fluid binary mixtures under an external temperature gradient, possible systems come naturally in two groups. The first consists of room-temperature mixtures, e.g., of two alcohols or alcohol and water; the second group has only one member, ^{3}He-^{4}He normal-fluid solutions. Both groups have advantages and disadvantages from an experimental as well as a theoretical point of view, and as we shall see they are complementary in many respects. In ^{3}He-^{4}He mixtures[27,28] the disadvantage in studying convection is an obvious one: Up to now there exists no technique which allows a visualization of the flow pattern above the onset of convection. Therefore the experiments are confined to measurements of the heat current (i.e., the Nusselt number), which can be done with very high accuracy.[29] On the other hand, ^{3}He-^{4}He mixtures offer the unique advantage that the dimensionless parameters such as the separation ratio and the Schmidt and Prandtl numbers can be varied over orders of magnitude. This is, in particular, true in the vicinity of the λ line (close to the superfluid transition) and in the neighborhood of the tricritical point (close to the two-phase region). In the ^{3}He concentration range between 40% and 50% and at temperatures of order 0.1–0.2 K from the λ line, the typical value for $L^{-1} = \kappa / D$ is 3. This yields for the separation ratio at the polycritical point, $\psi_{\text{pc}} \simeq -0.1$, a value which may be reached near the superfluid transition by varying concentration and temperature. Moreover, there is an additional advantage in this region that the value of ψ_{pc} is reasonably well separated in parameter space from the "nonequilibrium tricritical point" for the stationary bifurcation at which the cubic term in the amplitude equation undergoes a change in sign.[11] For this point one obtains the value $\psi_t \simeq -0.07$. The second interesting region is the vicinity of the thermodynamic tricritical point. There, ψ can be easily varied from -0.01 to about -1 in a very narrow temperature range, thus including the polycritical value of ψ ($\psi_{pc} \simeq -0.01$ for $\kappa/D \simeq 10$). In the region of parameter space where the amplitude equation derived above predicts a stable Hopf bifurcation (region IIb of Fig. 1) we predict oscillations in the Nusselt number with frequency $2\omega_o$ and we further predict that this frequency goes to zero at the polycritical point and along the line L_1.

For room-temperature systems[20,21] the situation is completely different. One can observe the flow pattern using, for example, a shadowgraph technique,[30] for any given pair of values of the separation ratio and the applied temperature gradient. In addition, it is simpler to measure the time dependence of the flow using a visualization technique or by local temperature measurements at several points in the fluid. Spatial correlations can be determined in these room-temperature systems by using laser Doppler velocimetry, a technique not available in low-temperature convection. The disadvantage common to all room-temperature systems is the close proximity of the polycritical point ($\psi_{pc} \approx -10^{-2}$ for a typical alcohol-alcohol mixture) with the nonequilibrium tricritical point for the stationary instability ($\psi_t \approx -10^{-3}$). This means that it will be necessary in experiments to control both the temperature gradient and the separation ratio rather precisely in order to ensure reproducible results and to avoid confusing the different dynamical behaviors near these two points. Given the accuracy of the experimental techniques available at present and the fact that one can combine the results obtained by various methods, it should nevertheless be possible to obtain some information on the dynamic behavior in the vicinity of ψ_{pc}.

Let us compare the onset of convection in the vicinity of the polycritical point in a binary fluid mixture (with and without a porous medium) with other systems in which codimension-2 bifurcations could occur or have been studied. In a simple fluid one can clearly not have a codimension-2 bifurcation near onset, since the first instability is always stationary.[25] The system which has attracted most theoretical attention so far is the thermohaline problem,[8,3,7] i.e., the onset of convection in a layer of salt water. As mentioned in the Introduction, there is unfortunately one important experimental problem: To study codimension-2 bifurcations in the thermohaline problem it would be necessary to vary salinity and temperature *gradients* independently, and it appears quite difficult to do this experimentally, due to the coupling proportional to k_T which exists between these gradients in equilibrium mixtures.[21]

Another candidate for a codimension-2 bifurcation is a simple conducting fluid in an external magnetic field which serves as the second parameter. Although some model calculations[8] have been carried out on this problem, no experiments have been published so far, to our knowledge. (Mercury in a magnetic field appears to be a promising candidate.[31])

Very recently Andereck *et al.*[32] have reported experiments on the behavior of a simple fluid between corotating and counterrotating cylinders. The observations included several types of codimension-2 bifurcations, but no theoretical interpretation of the results has been attempted so far.

The experimental systems for which most data have been accumulated so far in the vicinity of a codimension-2 bifurcation in convection are nematic liquid crystals.[14,33–35] As in binary fluid mixtures one has the possibility of an oscillatory instability. The role of concentration is taken over by the fluctuations of the nematic director, the variable which characterizes the spontaneously broken rotational symmetry in these liquid crystals (the center of mass of the molecules shows no long-range order). As a convenient second parameter in nematics one has an external magnetic field which couples to the elastic free energy and can thus be used as a lever to change the critical Rayleigh number and the frequency at onset. The codimension-2 point and its vicinity have so far been studied only experimentally. Theories concentrated on the derivation of amplitude equations for the stationary and the oscillatory instability separately. Dubois-Violette and Rothen[36] found a stationary instability in the planar configuration in MBBA [N-(*p*-methoxybenzylidene)-*p*-butylaniline] with a forward bifurcation, and Dubois-Violette and Gabay[37] reported for the same compound in the homotropic configuration an inverse oscillatory bifurcation. The only experiments performed for MBBA have been carried out by Guyon, Pieranski, and Salan[35] and they seem to indicate an inverse Hopf bifurcation triggering a finite amplitude instability. The vicinity of the codimension-2 point was not studied in detail in these early reports. More recently, this question was addressed experimentally in a series of papers by Otnes and Riste.[14] The investigated material in their case was PAA (*p*-azoxyanisole) and they used neutron scattering to detect changes in the order parameter. In a small aspect ratio cell with a small magnetic field they found a forward Hopf bifurcation with sustained nonlinear oscillations, and at high fields they observed a stationary instability which was accompanied by spatially irregular behavior and possibly a strange attractor. At higher temperature gradients they found multistability for the oscillations and in addition they observed hysteresis. The experimental behavior for small temperature gradients resembles very closely what we have found in binary fluid mixtures: a forward Hopf bifurcation competes with an inverse stationary bifurcation. We mention in passing that it is possible to derive the structure of the amplitude equation near the polycritical point in nematics as well, and we have found very recently[38] that this equation is identical with that for the binary-fluid-mixture case. The situation differs only insofar as f_1 and f_2 are more complicated expressions which can probably assume either sign and thus allow for a rich variety of behavior which varies from one compound to the next (e.g., from MBBA to PAA).

In conclusion, it is our belief that binary fluid mixtures (with and without a porous medium) and nematic liquid crystals are the best candidates for study, since they are experimentally accessible and allow a theoretical description of the behavior close to the intersection of a stationary and an oscillatory instability in convection.

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APPENDIX A: DERIVATION OF THE AMPLITUDE EQUATION IN A POROUS MEDIUM

1. Derivation of Eq. (2.13) from the amplitude equations for the stationary instability and the oscillatory instability

For the stationary instability, we derived previously[11] the amplitude equation

$$\tau_s \dot{w}_s = \epsilon_s w_s - g_s w_s^3 , \tag{A1}$$

with

$$\tau_s = \frac{1}{2\pi^2}\left[1 + L^{-2}\frac{\psi R_{cs}}{4\pi^2}\right] , \tag{A2}$$

$$\epsilon_s = (R - R_{cs})/R_{cs} , \tag{A3}$$

and

$$g_s = \frac{1}{16\pi^2}\left[1 + \frac{R_{cs}\psi}{4\pi^2 L^2}(1 + L^{-1})\right] . \tag{A4}$$

The function $w_s = \frac{1}{2}W$ is related to the velocity by $w = 2w_s \sin(\pi z)\cos(\pi x)$. For the oscillatory instability we found, with

$$w = (w_o e^{i\omega_o t} + w_o^* e^{-i\omega_o t})\sin(\pi z)\cos(\pi x) , \tag{A5}$$

the complex "Ginzburg-Landau"-type equation

$$\tau_o \dot{w}_o = \epsilon_o w_o - g_o \mid w_o \mid^2 w_o , \tag{A6}$$

where

$$\tau_o = \frac{2i\omega_o}{(i\omega_o + 2\pi^2)(i\omega_o + 2\pi^2 L)} , \tag{A7}$$

$$\epsilon_o = (R - R_{co})/R_{co} , \tag{A8}$$

and

$$g_o = \frac{\pi^2}{2(4\pi^4 + \omega_o^2)}\left[1 + \frac{2\pi^2 - i\omega_o}{2(2\pi^2 + i\omega_o)} - \frac{\psi R_{co}(2\pi^2 - i\omega_o)}{4(2\pi^2 + i\omega_o)(2\pi^2 L + i\omega_o)} + \frac{4\pi^4 + \omega_o^2}{2\pi^2 L(2\pi^2 L + i\omega_o)} - \frac{R_{co}}{2L(2\pi^2 L + i\omega_o)}\right.$$
$$\left. + \frac{4\pi^4 + \omega_o^2}{2(2\pi^2 + L + i\omega_o)^2} - \frac{R_{co}(2\pi^2 - i\omega_o)}{4(2\pi^2 L + i\omega_o)^2} + \frac{\psi\pi^2 L R_{co}(2\pi^2 - i\omega_o)}{2(2\pi^2 + i\omega_o)(2\pi^2 L + i\omega_o)^2}\right] . \tag{A9}$$

To determine f_1 and f_2 in Eq. (2.13) from Eqs. (A1)—(A9) it is useful to keep in mind the situation sketched in Fig. 1, showing the intersection of the lines for the stationary and oscillatory instabilities. At point B we have $\beta > 0$ and $\beta \ll 1$ because B was chosen to lie close to the line $\beta = 0$. Furthermore, we can easily choose B so as to satisfy the inequalities $\alpha < 0, \mid\alpha\mid \gg \beta$. At B, $W^2 \approx O(\beta) \ll \mid\alpha\mid$, so that it is possible to neglect the term containing f_2 in (2.13), which becomes

$$-\alpha\dot{W} = \beta W + f_1 W^3 , \tag{A10}$$

where according to (2.6) and (A5) $W = w_o \exp(i\omega_o t) + \text{c.c.}$ Comparing Eq. (A10) with Eqs. (A1)—(A4) we read off immediately

$$f_1 = -4\pi^4 g_s L . \tag{A11}$$

If we now let B go to the polycritical point along R_{cs} we obtain for f_1 using Eq. (2.11)

$$f_1 = \frac{\pi^2}{4} . \tag{A12}$$

Next we perform a similar analysis for point A near the oscillatory line. We have chosen A to satisfy $\beta < 0$ and $0 < \alpha \ll \mid\beta\mid$. Using $\alpha \equiv \eta^2$ as a small parameter we carry out a perturbation expansion of Eq. (2.13) using $\tau = \eta^2 t$ as a slow time scale and t_1 as a fast time scale. Inserting the expansions

$$W = \eta W_1(t_2, \tau) + \eta^3 W_3(t_1, \tau) + \cdots \tag{A13}$$

and

$$\partial_t = \partial_{t_1} + \eta^2 \partial_\tau = \partial_1 + \eta^2 \partial_\tau \tag{A14}$$

into Eq. (2.13) we obtain to order η

$$(\partial_1^2 + \omega_o^2) W_1 = 0 , \tag{A15}$$

with $\beta = -\omega_o^2 + O(\eta^2)$, and to order η^3

$$(\partial_1^2 + \omega_o^2) W_3 + (2\partial_1\partial_\tau - \partial_1 + f_2 W_1^2 \partial_1 - f_1 W_1^2) W_1 = 0 . \tag{A16}$$

Using the solution of Eq. (A15),

$$W_1 = \overline{W}_1(\tau) e^{i\omega_o t} + \overline{W}_1^*(\tau) e^{-i\omega_o t} , \tag{A17}$$

we get for the solvability condition for the coefficient of $\exp(i\omega_o t)$ in (A16)

$$\dot{\overline{W}}_1 = \tfrac{1}{2}(1 + i\omega_o)\overline{W}_1 - \tfrac{1}{2}(f_2 + 3if_1/\omega_o) \mid \overline{W}_1 \mid^2 \overline{W}_1 . \tag{A18}$$

In order to identify the coefficients in (A18) we must compare with (A6) in the limit $\omega_0 \to 0$. A lengthy but straightforward expansion of (A7) and (A9) then leads to a confirmation of (A12) and to the further result

$$f_2 = \tfrac{1}{4}(1+L^{-1}) \, . \tag{A19}$$

2. Derivation of the amplitude equation from the nonlinear basic equations

We now wish to present the alternative derivation starting from the nonlinear equations for the deviations from the heat-conduction state, Eqs. (2.1)–(2.3), which we rewrite in the form

$$\mathscr{L}\xi = -\partial_t \mathscr{L}_1 \xi + \mathscr{L}_2 \xi = \mathscr{N}(\xi) \, , \tag{A20a}$$

where

$$\mathscr{L}_1 = \begin{bmatrix} \dfrac{-K}{l^2 \epsilon \nu L} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \mathscr{L}_2 = \begin{bmatrix} \Delta & -\Delta_2 & -\psi\Delta_2 \\ R & \Delta & 0 \\ R & -L\Delta & L\Delta \end{bmatrix} , \tag{A20b}$$

$$\mathscr{N}(\xi) = \begin{bmatrix} 0 \\ (\vec{\mathrm{v}}\cdot\vec{\nabla})\theta \\ (\vec{\mathrm{v}}\cdot\vec{\nabla})c \end{bmatrix}, \quad \xi = \begin{bmatrix} w \\ \theta \\ c \end{bmatrix} . \tag{A20c}$$

Linearizing Eq. (A20) and taking a Fourier transform we obtain from the condition

$$\mathscr{L}_2 \xi_1^0 = 0 \tag{A21}$$

a first right zero eigenvector (of $\mathscr{L}$)

$$\xi_1^0 = \begin{bmatrix} 1 \\ R/k^2 \\ R/k^2(1+L^{-1}) \end{bmatrix} \sin(\pi z)\cos(\pi x) \, , \tag{A22}$$

where, in Eq. (A22) as in the following, R and ψ are always evaluated at the polycritical point (2.11) and $k^2 = k_\parallel^2 + k_\perp^2$. Since Eq. (2.7) has a double zero eigenvalue at the intersection of the lines for the stationary and oscillatory instabilities we must determine a second right eigenvector which can be evaluated from

$$\mathscr{L}_2 \xi_2^0 = \mathscr{L}_1 \xi_1^0 \, . \tag{A23}$$

We are left with

$$\xi_2^0 = \begin{bmatrix} 0 \\ -R/k^4 \\ R/\psi k^4 \end{bmatrix} \sin(\pi z)\cos(\pi x) + \zeta \xi_1^0 \, , \tag{A24}$$

with ζ an arbitrary constant [in view of (A21)] which we take to be zero for convenience. Proceeding in an analogous way for the left eigenvectors we obtain

$$\xi_1^{0l} = \begin{bmatrix} 1 \\ (k_\perp^2/k^2)(1+\psi) \\ \psi k_\perp^2/k^2 L \end{bmatrix} \sin(\pi z)\cos(\pi x) \tag{A25}$$

and

$$\xi_2^{0l} = \begin{bmatrix} 0 \\ \psi k_\perp^2/k^4 L^2 \\ -\psi k_\perp^2/k^4 L^2 \end{bmatrix} \sin(\pi z)\cos(\pi x) + \tilde{\zeta} \xi_1^{0l} \, , \tag{A26}$$

with $\tilde{\zeta}$ again arbitrary at this stage. After these preliminary remarks about the linear analysis on the polycritical surface we are prepared to carry out a nonlinear analysis.

As has already been mentioned above, one expects an equation of the form

$$\ddot{W} + \alpha \dot{W} + \beta W = f(W, \dot{W}) \, ,$$

where $f(W,\dot{W})$ contains all nonlinear terms, the lowest of which are cubic by symmetry. We make the ansatz

$$f = f_1 W^3 + f_2 W^2 \dot{W} + f_3 \dot{W}^2 W + f_4 \dot{W}^3 \, . \tag{A27}$$

Now it is fairly clear how to proceed: one tries a power expansion of the solutions of the full nonlinear equations, generalizing a technique of the Stuart type,[39]

$$\xi = \sum_{i,j} \xi_{ij} (W_1)^i (W_2)^j + \xi_1^0 W_1 + \xi_1^0 W_2 \, , \tag{A28}$$

where $W_1 \equiv W$ and $W_2 \equiv \dot{W}$, and each ξ_{ij} is a vector. If such an expansion is valid one can expect that only ξ_{02}, ξ_{11}, and ξ_{20} need to be determined to derive a cubic equation for W. Using

$$\partial_t \xi = \frac{\partial \xi}{\partial W_1} \dot{W}_1 + \frac{\partial \xi}{\partial W_2} \dot{W}_2 \, , \tag{A29}$$

we can rewrite Eq. (A20) as

$$-\mathscr{L}_1 W_2 \frac{\partial \xi}{\partial W_1} - \mathscr{L}_1 \frac{\partial \xi}{\partial W_2}(-\alpha W_2 - \beta W_2 + f) + \mathscr{L}_2 \xi = \mathscr{N}(\xi) \, . \tag{A30}$$

All that is left to do is to insert Eqs. (A27) and (A28) into Eq. (A30) and to determine the coefficients $f_1, \dots, f_4$ self-consistently in the reduced perturbation scheme.

To first order in the iteration scheme we get for the nonlinearities

$$\mathscr{N}(\xi_1^0, \xi_1^0) = \begin{bmatrix} 0 \\ \pi R/2k^2 \\ (\pi R/2k^2)(1+L^{-1}) \end{bmatrix} \sin(2\pi z) \, , \tag{A31}$$

$$\mathscr{N}(\xi_1^0, \xi_2^0) = \begin{bmatrix} 0 \\ -\pi R/2k^4 \\ \pi R/2\psi k^4 \end{bmatrix} \sin(2\pi z) \, , \tag{A32}$$

and

$$\mathscr{N}(\xi_2^0, \xi_2^0) = 0 \, . \tag{A33}$$

The coefficients ξ_{20}, ξ_{11}, and ξ_{02} can then easily be determined [using (A30)]:

$$\begin{aligned} &\mathscr{L}_2 \xi_{20} = \mathscr{N}(\xi_1^0, \xi_1^0) \, , \\ &\mathscr{L}_2 \xi_{11} = \mathscr{N}(\xi_1^0, \xi_2^0) + 2\mathscr{L}_1 \xi_{20} \, , \\ &\mathscr{L}_2 \xi_{02} = 2\mathscr{L}_1 \xi_{11} \, , \end{aligned} \tag{A34}$$

leaving us with

$$\xi_{20}=\begin{bmatrix}0\\ -R/16\pi^3\\ (-R/16\pi^3)(1+L^{-1}+L^{-2})\end{bmatrix}\sin(2\pi z)\ , \tag{A35}$$

$$\xi_{11}=\begin{bmatrix}0\\ R/16\pi^5\\ (R/16\pi^5)[1-(L\psi)^{-1}]\end{bmatrix}\sin(2\pi z)\ , \tag{A36}$$

and

$$\xi_{02}=\begin{bmatrix}0\\ -R/32\pi^7\\ (-R/32\pi^7)\{1+[1-(L\psi)^{-1}]/L\}\end{bmatrix}\sin(2\pi z)\ . \tag{A37}$$

Due to the up-down symmetry, all orthogonality conditions occurring in second order are satisfied trivially, thus justifying again the omission of terms quadratic in W and/or $\dot{W}$ in the ansatz for f. To determine $f_1,\ldots,f_4$ we must evaluate the orthogonality conditions (because we have a larger critical space there are four such conditions here instead of one for the Rayleigh-Bénard instability in simple fluids). In detail, we have from Eq. (A20)

$$\mathcal{L}_2\xi_{30}=\mathcal{N}(\xi_{20},\xi_1^0)+f_1\mathcal{L}_1\xi_2^0\ , \tag{A38}$$

$$\mathcal{L}_2\xi_{21}=\mathcal{N}(\xi_{11},\xi_1^0)+f_2\mathcal{L}_1\xi_2^0+3\mathcal{L}_1\xi_{30}\ , \tag{A39}$$

$$\mathcal{L}_2\xi_{12}=\mathcal{N}(\xi_{02},\xi_1^0)+f_3\mathcal{L}_1\xi_2^0+2\mathcal{L}_1\xi_{21}\ , \tag{A40}$$

$$\mathcal{L}_2\xi_{03}=f_4\mathcal{L}_1\xi_2^0+\mathcal{L}_1\xi_{12}\ . \tag{A41}$$

From Eq. (A38) we immediately find f_1, using the first of the orthogonality conditions:

$$f_1=-\frac{(\xi_1^{0l},\mathcal{N}(\xi_{20},\xi_1^0))}{(\xi_1^{0l},\mathcal{L}_1\xi_2^0)}\ , \tag{A42}$$

where we have introduced the inner product as an integral over the fluid layer

$$(A,B)=\int dx\,dz\,A(x,z)B(x,z)\ .$$

From Eqs. (A24), (A25), and (A35) we have immediately

$$f_1=\frac{\pi^2}{4}\ , \tag{A43}$$

as in Eq. (A12). Rewriting Eq. (A38) as

$$\xi_{30}=\mathcal{L}_2^{-1}[\mathcal{N}(\xi_{20},\xi_1^0)+f_1\mathcal{L}_1\xi_2^0]\ , \tag{A44}$$

we obtain from Eq. (A44)

$$(\mathcal{L}_1\xi_{30},\xi_1^{0l})=(\mathcal{N}(\xi_{20},\xi_1^0)+f_1\mathcal{L}_1\xi_2^0,(\mathcal{L}_2^+)^{-1}\mathcal{L}_1^+\xi_{10}^l) \tag{A45}$$

$$=(\mathcal{N}(\xi_{20},\xi_1^0)+f_1\mathcal{L}_1\xi_2^0,\xi_2^{0l}) \tag{A46}$$

$$=(\mathcal{N}(\xi_{20},\xi_1^0),\xi_2^{0l})\ . \tag{A47}$$

To arrive at Eq. (A47) we have chosen $(\xi_2^0,\xi_2^{0l})=0$ which fixes $\tilde{\zeta}$ in Eq. (A26). Combining Eqs. (A47) and (A39) we arrive at

$$f_2=-\frac{(\mathcal{N}(\xi_{11},\xi_1^0),\xi_1^{0l})-3(\mathcal{N}(\xi_{20},\xi_1^0),\xi_2^{0l})}{(\mathcal{L}_2\xi_2^0,\xi_1^{0l})}\ . \tag{A48}$$

Inserting ξ_{11}, ξ_1^{0l}, ξ_2^{0l}, and ξ_{20} into Eq. (A48) we find after a straightforward calculation

$$f_2=\tfrac{1}{4}(1+L^{-1})\ , \tag{A49}$$

a result which agrees with Eq. (A19) above. We are thus left with the determination of f_3 and f_4. From Eq. (A38) it is clear that ξ_{30} is given by a special solution of the inhomogeneous equation plus the general solution of the homogeneous equation $c\xi_0^1$ with c an arbitrary constant. The same type of argument can be used for the general solution of Eq. (A39). From Eq. (A40) it follows that the compatibility condition for this equation pins down only an arbitrary linear combination of f_3 and the arbitrary constant c entering ξ_{30}. Accordingly, we may choose this constant so that $f_3\equiv 0$. The same argument applied to the orthogonality condition for Eq. (A41) yields $f_4\equiv 0$, which completes our derivation of the amplitude equation (2.13). A more general derivation of the form of this equation is given in Appendix C.

APPENDIX B: DERIVATION OF THE AMPLITUDE EQUATION FOR BULK MIXTURES

To derive Eqs. (2.28) and (2.29) we use the direct method, i.e., we obtain the amplitude equation from the basic nonlinear equations. Since the steps are similar to those in Appendix A 2 we shall use the same notation and list the results only. For $\mathcal{L}_1$ and $\mathcal{L}_2$ we have

$$\mathcal{L}_1=\begin{bmatrix}-P^{-1}\Delta & \Delta & 0\\ 0 & 0 & 0\\ 0 & 0 & 1\end{bmatrix}\ , \tag{B1}$$

$$\mathcal{L}_2=\begin{bmatrix}-\Delta^2 & -\Delta_2 & -\Delta_2\psi\\ R & \Delta & 0\\ R & -L\Delta & L\Delta\end{bmatrix}\ . \tag{B2}$$

For the first zero eigenvector we have then

$$\xi_1^0=\begin{bmatrix}1\\ R/k^2\\ (R/k^2)(1+L^{-1})\end{bmatrix}\ , \tag{B3}$$

with $k^2=k_{\parallel}^2+k_{\perp c}^2$, and ξ_2^0 reads

$$\xi_2^0=\begin{bmatrix}0\\ -R/k^4\\ (R/k^2-k^2/k_\perp^2P)/\psi\end{bmatrix}+\zeta\xi_1^0\ . \tag{B4}$$

For the left eigenvectors ξ_1^{0l} and ξ_2^{0l} we find

$$\xi_1^{0l}=\begin{bmatrix}1\\ (k_\perp^2/k^2)(1+\psi)\\ \psi k_\perp^2/k^2L\end{bmatrix} \tag{B5}$$

and

$$\xi_2^{0l}=\begin{bmatrix}0\\-(k_\perp^2/k^4)(1+\psi+\psi/L)\\ \psi k_\perp^2/k^2L^2\end{bmatrix}+\hat{\zeta}\xi_1^{0l}\,. \tag{B6}$$

For the nonlinearities we get

$$\mathcal{N}(\xi_1^0,\xi_1^0)=\begin{bmatrix}0\\ \pi k_\perp R/2k^2\\ (\pi R k_\perp/2k^2)(1+L^{-1})\end{bmatrix}, \tag{B7}$$

$$\mathcal{N}(\xi_1^0,\xi_2^0)=\begin{bmatrix}0\\ -\pi k_\perp R/2k^4\\ (-\pi R/2k^4)(1+L^{-1}+L^{-2})\end{bmatrix}. \tag{B8}$$

In the next step we determine, as in Appendix A 2, the quantities ξ_{20} and ξ_{11}:

$$\xi_{20}=\begin{bmatrix}0\\ -k_\perp R/8\pi k^2\\ (-k_\perp R/8\pi k^2)(1+L^{-1}+L^{-2})\end{bmatrix}\sin(2\pi z)\,, \tag{B9}$$

$$\xi_{11}=\begin{bmatrix}0\\ 7k_\perp R/72\pi^5\\ (Rk_\perp/\pi^5)[\frac{7}{72}+(1+L^{-1}+L^{-2})/24L-(18L\psi)^{-1}]-3k_\perp/8\pi L\psi P\end{bmatrix} \tag{B10}$$

In the final step we determine f'_1 and f'_2, and we obtain

$$f'_1=\tfrac{3}{32}\pi^4 \tag{B11}$$

and

$$f'_2=(7\pi^2/64)[1+(1+P)L^{-2}(1+L^{-1}+P/L)^{-1}]\,, \tag{B12}$$

as in Eqs. (2.30) and (2.31) of the text.

APPENDIX C: CANONICAL FORMS FOR THE AMPLITUDE EQUATION FOR CODIMENSION-2 AND -3 BIFURCATIONS

In Appendix A 2 we have given a detailed argument why terms proportional to $\dot{W}^2W$ and $\dot{W}^3$ may be omitted from Eq. (2.13). Here, we wish to discuss this point from a more general point of view.[40] Accordingly we place ourselves on the polycritical surface $\alpha=\beta=0$ and start from the most general form for a system having "up-down" symmetry,

$$\ddot{W}'=f_1W'^3-f_2W'^2\dot{W}'+f_3\dot{W}'^2W'+f_4\dot{W}'^3\,, \tag{C1}$$

for an amplitude function W' which is related to W by a nonlinear transformation

$$W'=W+d_1W^3+d_2W^2\dot{W}+d_3W\dot{W}^2+d_4\dot{W}^3\,. \tag{C2}$$

From (C2) it follows immediately that

$$\dot{W}'=\dot{W}+d_13W^2\dot{W}+d_2(2\dot{W}^2W+W^2\ddot{W})$$
$$+d_3(\dot{W}^3+2W\dot{W}\ddot{W})+3d_4\dot{W}^2\ddot{W}\,, \tag{C3}$$

and

$$\ddot{W}'=\ddot{W}+3d_1(W^2\ddot{W}+2\dot{W}^2W)$$
$$+d_2(2\dot{W}^3+6W\dot{W}\ddot{W}+W^2\dddot{W})$$
$$+d_3(5\dot{W}^2\ddot{W}+2W\ddot{W}^2+2W\dot{W}\dddot{W})$$
$$+3d_4(\dddot{W}\dot{W}^2+2\dot{W}\ddot{W}^2)\,. \tag{C4}$$

Since according to Eqs. (C1) and (C2) $\ddot{W}$ and $\dddot{W}$ are at least of order W^3, $W^2\dot{W}$, or $W\dot{W}^2$, the terms containing these may be dropped from the right-hand side of Eq. (C4), which simplifies to give to the order we are interested in

$$\ddot{W}'=\ddot{W}+6d_1\dot{W}^2W+2d_2\dot{W}^3\,. \tag{C5}$$

In comparing Eq. (C5) with Eq. (C1) it is obvious that the terms in (C1) proportional to $\dot{W}^3$ and $\dot{W}^2W$ can be removed by appropriate choice of the d_i, whereas terms proportional to W^3 and $W^2\dot{W}$ will remain. Thus we end up with the "canonical" form for the equation on the polycritical surface associated with a codimension-2 bifurcation,

$$\ddot{W}=f_1W^3-f_2W^2\dot{W}\,. \tag{C6}$$

For codimension-3 bifurcations which were studied by Arnéodo, Coullet, and Spiegel,[41] a similar technique can be applied. The most general equation reads in this case

$$\dddot{W}'=c_1W'^3+c_2W'^2\dot{W}'+c_3W'\dot{W}'^2$$
$$+c_4W'^2\ddot{W}'+c_5W'\ddot{W}'^2+c_6\ddot{W}'^3+c_7\dot{W}'\ddot{W}'^2$$
$$+c_8\dot{W}'^2\ddot{W}'+c_9W'\dot{W}'\ddot{W}'+c_{10}\dot{W}'^3\,. \tag{C7}$$

Making the nonlinear transformation

$$W' = W + d_0 W^3 + d_1 W^2 \dot{W} + d_2 W \dot{W}^2 + d_3 \dot{W}^3 + d_4 W^2 \ddot{W} + d_5 W \ddot{W}^2 + d_6 \ddot{W}^3 + d_7 \dot{W} \ddot{W}^2 + d_8 \dot{W}^2 \ddot{W} + d_9 W \dot{W} \ddot{W} , \tag{C8}$$

one arrives to the order we are investigating here, using similar arguments as above, at the following expression for $\dddot{W}'$:

$$\dddot{W}' = \dddot{W} + 3d_2(6W\dot{W}\ddot{W} + 2\dot{W}^3) + d_3(12\dot{W}^2\ddot{W} + 6W\ddot{W}^2) + 12d_4 \dot{W}\ddot{W}^2 + 12d_5 \ddot{W}^3 + 6d_6 \dot{W}\ddot{W}^2 + 3d_9 \ddot{W}^3 . \tag{C9}$$

By comparing Eqs. (C9) and (C7) we see immediately that the terms proportional to $\ddot{W}^3$ and $\dot{W}\ddot{W}^2$ can be removed by a nonlinear transformation. In addition we find a rather surprising result: there is a choice as to which other terms are removed. One of the following four combinations of nonlinearities,

$$\{\dot{W}\ddot{W}W, \dot{W}^2\ddot{W}\},\ \{\dot{W}\ddot{W}W, W\ddot{W}^2\},\ \{\dot{W}^3, \dot{W}^2\ddot{W}\},\ \{\dot{W}^3, W\ddot{W}^2\} , \tag{C10}$$

can be removed, but not the other three. From this result we can infer immediately that Arnéodo, Coullet, and Spiegel[41] in their Eq. (31) have chosen to remove the first pair, and that they have retained the other three, but one could have selected any other of the combinations listed above—a choice which does not exist for the case of codimension 2.

APPENDIX D: LIMIT CYCLE SOLUTION OF EQ. (2.13)

In this section we display the periodic solution of Eq. (2.13) which exists in sector II of Fig. 1, using an argument which has been given by a number of authors.[3,4,6,7,42] In the limit $\alpha,\beta \to 0$ we introduce the scaling

$$W(t) = \epsilon x_1(\tau) , \tag{D1a}$$

$$\beta = \epsilon^2 \lambda_1 , \tag{D1b}$$

$$\alpha = \epsilon^2 \lambda_2 , \tag{D1c}$$

$$t = \epsilon^{-1}\tau , \tag{D1d}$$

$$\dot{W} = \partial_t W = \epsilon^2 x_2(\tau) = \epsilon^2 \partial_\tau x_1 \tag{D1e}$$

and rewrite Eq. (2.13) as

$$\dot{x}_1 = x_2 , \tag{D2}$$

$$\dot{x}_2 = \lambda_1 x_1 + f_1 x_1^3 + \epsilon(\lambda_2 x_2 - f_2 x_1^2 x_2) . \tag{D3}$$

For $\epsilon = 0$, Eq. (D3) is a Hamiltonian equation with energy

$$H = \tfrac{1}{2}x_2^2 - \tfrac{1}{2}\lambda_1 x_1^2 - \tfrac{1}{4}f_1 x_1^4 . \tag{D4}$$

To zeroth order in ϵ the solutions of (D4) are therefore lines of constant energy $H(x_1,x_2) = \text{const}$. Since areas are conserved by a Hamiltonian flow the change of phase space area of the motion given by (D3) depends only on the term proportional to ϵ. It is obtained by integrating the divergence of the vector flow $\partial \dot{x}_1/\partial x_1 + \partial \dot{x}_2/\partial x_2$ around the orbit. Since we are dealing with a limit cycle the area A does not change with time (even for finite ϵ), so we have the condition

$$\frac{dA}{d\tau} = \epsilon \int dx_1 dx_2 (\lambda_2 - f_2 x_1^2) = 0 . \tag{D5}$$

We wish to find the point in (λ_1,λ_2) space at which the limit cycle $x_1(\tau), x_2(\tau)$ touches the fixed points at $x_1 = \pm(-\lambda_1/f_1)^{1/2}, x_2 = 0$. At these points the energy (D4) is $H = \frac{1}{4}\lambda_1^2/f_1 + O(\epsilon)$. To lowest order in ϵ we may thus evaluate Eq. (D5) in a limit cycle for which

$$H(x_1,x_2) = \tfrac{1}{2}x_2^2 - \tfrac{1}{2}\lambda_1 x_1^2 - \tfrac{1}{4}f_1 x_1^4 = \tfrac{1}{4}\lambda_1^2/f_1 . \tag{D6}$$

Inserting Eq. (D6) into Eq. (D5) we obtain

$$0 = \int_{-(-\lambda_1/f_1)^{1/2}}^{(-\lambda_1/f_1)^{1/2}} dx_1 [(\lambda_2 - f_2 x_1^2)(f_1/2)^{1/2}(x_1^2 + \lambda_1/f_1)] , \tag{D7}$$

which may be evaluated to yield

$$\lambda_2/\lambda_1 = -f_2/5f_1 , \tag{D8}$$

leading to Eq. (2.35) of the text. The stability of the limit cycle within sector IIb, where it exists, may also be verified to lowest order in ϵ, using the Hamiltonian limit.

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