Tricritical behavior in stationary double diffusive convection with cross diffusion

R. Becerril
Instituto de Física y Matemáticas.
Universidad Michoacana de San Nicolás de Hidalgo
Apartado Postal 2-82, 58040 Morelia, Michoacán, México

J. Mendoza
Escuela de Ciencias Fisico Matemáticas,
Universidad Michoacana de San Nicolás de Hidalgo
Apartado Postal 2-071, 58060, Morelia, Michoacán, México

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We derive an amplitude equation for the stationary instability for the isothermal double diffusive system with cross diffusion. A quintic term is computed in terms of rescaled cross-diffusion constants. This quintic term is stabilizing in the vicinity of the tricritical point. The existence of a tricritical point indicates the presence of a hysteresis loop whose parameters are also presented.

Keywords: Hydrodynamic stability; convection.

Derivamos una ecuación de amplitud para la inestabilidad estacionaria del sistema isotérmico doble difusivo con difusión cruzada. Se calcula un término a quinto orden en términos de las constantes re-escaladas de difusión cruzada. Este término es estabilizador en la vecindad del punto tricrítico. La existencia de un punto tricrítico indica la presencia de un ciclo de histéresis, cuyos parámetros se presentan en esta carta.

Descriptores: Estabilidad hidrodinámica; convección.

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1. Introduction.

The study of the dynamics of fluid layers in which there are gradients of two properties with different molecular diffusivities is known as double diffusive convection. The components most commonly used in these systems have been heat and salinity [1]. However, experiments performed a few years ago by Predtechensky *et al.* are isothermal and convective flow is driven by imposed vertical concentration gradients of two species with different diffusion constants [2]. Convective motion takes place in a very thin Hele-Shaw cell of length L, and height d. In this system, a fixed concentration of fast diffusing species c_{f0} is imposed at the top of the Hele-Shaw cell (where the concentration of the slow diffusing species vanishes), and a fixed concentration of a slow diffusing species c_{s0} is imposed at the bottom of the cell (where the concentration of the fast species is zero). The novel cell employed by Predtechensky in the experiment provided these well defined boundary conditions for the concentrations at the top and bottom of the cell. The resultant density gradient for the slow diffusive species was stabilizing, while that for the fast diffusing species was destabilizing.

As in thermosolutal [1] and binary mixture convection [3], isothermal double diffusive convection exhibits a rich variety of bifurcation phenomena. Above a critical value of the solutal Rayleigh number for the fast diffusing solute R_f, stationary convection arises depending on the value of the solutal Rayleigh number for the slow diffusing solute R_s. The system also presents a codimension 2-point (CTP) bifurcation where the stationary and oscillatory instabilities collide. Along the stationary branch, there is a tricritical point (TCP). It has been shown that since the Lewis number can be varied between the values 0.1 to nearly 1, these two points can be made well separated in the Rayleigh number R_s [4]. Hence isothermal double diffusive convection in a Hele-Shaw cell is a promising system in which is feasible to experimentally investigate its dynamics near these points. This is not the case for other double diffusive systems at room temperature where these points are very close to each other in their corresponding parameter space [5].

It is known that amplitude equations can help us model the occurrence and evolution of instabilities near the onset of convection. An analysis of the bifurcations presented in this system was carried out recently [4]. Nonetheless, cross diffusion was neglected. A comparison between the measured traveling wave frequency ω_0 at the onset of convection and the predicted frequency of linear stability analysis without cross-diffusion was made in Ref. 2. This comparison confirmed qualitatively the square root dependence of ω_0 with R_s; however, there was a deviation between experimental and predicted frequency that may be due to cross diffusion effects, as Predtechensky suggested [2]. A study of how cross diffusion affects the onset of convection was recently made [6]. In particular the location of the tricritical point along the stationary branch was found in terms of rescaled cross-diffusion constants. The existence of a tricritical point indicates that there may be a hysteresis loop along the stationary branch [7]. In this letter, we derive a fifth order amplitude equation using a perturbation scheme. This allows us to pin down the sign and magnitude of this quintic term and to roughly determine the expected amount of hysteresis in the vicinity of the TCP. Thereby it is possible to predict the crossover from critical behavior ($A^2 \sim \epsilon$) to tricritical behavior ($A^2 \sim \epsilon^{1/2}$) with $\epsilon = (R_f - R_{fc}^{ss})/R_{fc}^{ss}$, a prediction that may be experimentally tested.

2. Equations of motion

The basic equations of motion are the continuity and the Navier-Stokes equation together with the diffusion equations for the concentrations. If $\omega/d << 1$ (where ω is the width of the cell) then the system can be considered 2-dimensional and the term $\nu \bigtriangledown^2 \vec{u}$ appearing in the Navier Stokes equation can be replaced by $-12\nu\vec{u}/w^2$ [8] (ν is the kinematic viscosity). The Rayleigh numbers for the slow ($i = s$) and for the fast ($i = f$) species are defined by $R_i = \alpha_i c_{i0} g d w^2/12\nu D_f$; where g is the acceleration due to gravity, and α_i the derivative of the logarithm of the density with respect to the concentration deviation c_i from the conduction profile. In the conducting state, the fluid is at rest, and the dimensional concentrations $\hat{c}_s$ and $\hat{c}_f$ depend linearly upon the dimensional vertical coordinate $\hat{z}$ as $\hat{c}_s = c_{s0}(1 - \hat{z}/d)$ and $\hat{c}_f = c_{f0}(\hat{z}/d)$. We non-dimensionalize the equations as in Ref. 9. The rescaled cross-diffusion constants are defined as $\tau_{ss} \equiv \tau = D_{ss}/D_{ff}$, $\tau_{fs} = D_{fs}\alpha_f/D_{ff}\alpha_s$, and $\tau_{sf} = D_{sf}\alpha_s/D_{ff}\alpha_f$, $\tau_{ff} = 1$. τ is the ordinary Lewis number, and D_{ij} are dimensional-diffusion constants. τ_{ij} describes the flow of the species "i" due to the gradient of the species "j". Even though there is an Onsager relation [10] between these off-diagonal elements, it seems there is no relevant thermodynamics derivative matrix [2]. As a result, these cross-diffusion terms were introduced as independent parameters in Ref. 2.

The typical value of the modified Schmidt number $\sigma = 12\nu d^2/D_{ff}w^2$ for the different cells and fluids used in Predtechensky *et al*'s is of the order 10^5 [2]. Therefore, we work with the hydrodynamic equations in the limit of σ going to infinity. The basic dimensionless nonlinear equations for the deviations from the conductive state in the Oberbeck-Boussinesq approximation with cross-diffusion terms read [9]

$$\begin{pmatrix} \bigtriangledown^2 & -\partial_x & -\partial_x \\ R_s\partial_x & -\tau\bigtriangledown^2+\partial_t & -\tau_{sf}\bigtriangledown^2 \\ -R_f\partial_x & -\tau_{fs}\bigtriangledown^2 & -\bigtriangledown^2+\partial_t \end{pmatrix}\vec{\xi}=\begin{pmatrix} 0 \\ J(\psi,c_s) \\ J(\psi,c_f) \end{pmatrix}, \quad (1)$$

where

$$\vec{\xi}(x,z) = \begin{pmatrix} \psi \\ c_s \\ c_f \end{pmatrix}, \quad (2)$$

ψ being the stream function, and c_s and c_f the deviations of the concentrations from the conductive profile. $J(f,g)$ is the Poisson bracket in the $x - z$ variables.

3. Linear stability analysis

The goal of linear stability theory is to ascertain when a given state of a system, in our case the conductive state, is unstable to a small perturbation $\vec{\xi}$. Linear stability analysis yields the conditions under which the system will undergo a transition to a convective state. We assume periodicity in the horizontal direction, and vertical velocity vanishes at the bottom $(z = 0)$ and the top $(z = 1)$ of the cell. Thus the eigenfunctions read

$$\vec{\xi}(x,z) = \frac{1}{2}\begin{pmatrix} iA\exp(-iqx) \\ B\exp(-iqx) \\ C\exp(-iqx) \end{pmatrix}\exp(\lambda t)\sin(\pi z) + c.c., \quad (3)$$

Using $\vec{\xi}$, linear stability analysis yields a dispersion relation from which the stationary and oscillatory critical Rayleigh numbers are found to be [6]

$$R_{fc}^{ss} = \frac{R_s(1-\tau_{fs})}{\tau-\tau_{sf}} + \frac{k^4(\tau-\tau_{sf}\tau_{fs})}{q^2(\tau-\tau_{sf})}, \quad (4)$$

$$R_{fc}^{osc} = R_s + \frac{k^4}{q^2}(1+\tau) \quad (5)$$

Here $k^2 = q^2 + \pi^2$. The critical wave number for both instabilities is $q = \pi$. The codimension-2 point bifurcation occurs where both instabilities collide. In parameter space (R_s, R_f), it is given by

$$R_s^{c2} = 4\pi^2\frac{\tau^2 - \tau_{sf}(1+\tau-\tau_{fs})}{1-\tau+\tau_{sf}-\tau_{fs}}, \quad (6)$$

$$R_f^{c2} = 4\pi^2\frac{1+\tau_{fs}(\tau_{sf}-\tau-1)}{1-\tau+\tau_{sf}-\tau_{fs}}. \quad (7)$$

The frequency at onset of the traveling waves is

$$\omega_o^2 = q^2R_s(1-\tau+\tau_{sf}-\tau_{fs}) + k^4\left(\tau_{sf}(1+\tau-\tau_{fs})-\tau^2\right). \quad (8)$$

which vanishes at the CTP as can be easily verified.

4. Amplitude equation

Amplitude equations are simplified mathematical models that describe the slow spatial and temporal variations of the original variables that characterize any given system near criticality. They help us to determine the nature of the different bifurcations and capture the essential dynamics near threshold. A set of third order amplitude equations corresponding to the stationary, oscillatory and codimension-2 point bifurcations was derived for isothermal double diffusive convection with cross diffusion terms included in Ref. 6. For the stationary instability this equation reads

$$\tau_0\partial_t A = \epsilon A - g_3 A|A|^2, \quad (9)$$

where $\epsilon = (R_{fc} - R_{fc}^{ss})/R_{fc}^{ss}$ is the parameter of the transition, and τ_0 is the relaxation time. If we write the complex amplitude of the stream function as $A = \mathcal{A}\exp(i\theta)$, we find that $d\theta/dt = 0$ and that $\mathcal{A}$ satisfies (9). By introducing $B(t) = \mathcal{A}^2(t)$, (9) acquires the form $dB/dt = aB + bB^2$ which is a Bernoulli's equation whose solution reads

$$B(t) = \frac{\epsilon B(0)\exp(2\epsilon t/\tau_0)}{B(0)g_3(\exp(2\epsilon t/\tau_0) - 1) + \epsilon}, \tag{10}$$

For $\epsilon < 0$, $B(t) \to 0$ as $t \to \infty$, but for $\epsilon > 0$, $B(t) = \mathcal{A}^2 \to \epsilon/g_3$ as $t \to \infty$. Therefore, when the asymptotic solution is not the trivial one, and $g_3 > 0$ a nontrivial stable solution exists for $\epsilon > 0$, and the bifurcation is called forwards or supercritical, whereas if $g_3 < 0$ a nontrivial unstable solution exists for $\epsilon < 0$ and the bifurcation is called backwards or subcritical. The point at which there is a shift from supercritical to subcritical bifurcation is known as the tricritical point. We actually do not need to know the explicit solution of (9) in order to find out that $\mathcal{A}^2 = \epsilon/g_3$ we just have to consider the steady state of (9), namely $\epsilon A - g_3 A|A|^2 = 0$. Thus nonlinear coefficient g_3 determines the bifurcation behavior of the system along the steady branch. In order to compute the coefficients g_3 and τ_0, we expand the fields and the Rayleigh number R_f in terms of a small parameter η as follows:

$$R_f = R_{fc}^{ss} + \eta^2 R_2^{ss} + \eta^4 R_4^{ss} + \ldots, \tag{11}$$

$$\vec{\xi} = \frac{1}{2}(\eta\vec{\xi}_1 A + \eta^2\vec{\xi}_2|A|^2 + \eta^3\vec{\xi}_3 A|A|^2 + \ldots + c.c.), \tag{12}$$

and replace ∂_t by $\eta^2\partial_T$. Inserting these expansions into Eq. (1), we find a series of linear equations at different orders in η whose integrability condition involves the solution of the adjoint of the linear operator [11]

$$\mathcal{L}_0 \equiv \begin{pmatrix} \nabla^2 & -\partial_x & -\partial_x \\ R_s\partial_x & -\tau\nabla^2 & -\tau_{sf}\nabla^2 \\ -R_{fc}^{ss}\partial_x & -\tau_{fs}\nabla^2 & -\nabla^2 \end{pmatrix}. \tag{13}$$

The equation at order η is $\mathcal{L}_0\vec{\xi}_1 = 0$ and has the solution $\vec{\xi}_1 \propto \exp(-i\pi x)\sin(\pi z)$. At second order we find that $\vec{\xi}_2 \propto \sin(2\pi z)$. To third order, the integrability condition yields Eq. (9), with the coefficients g_3 and τ_0 given by [6]

$$g_3 = \frac{1}{16(\tau - \tau_{sf})(\tau - \tau_{sf}\tau_{fs})}\big[R_s(1+\tau)(-1+\tau+\tau_{fs}-\tau_{sf}) + 4\pi^2[\tau^3 - \tau^2\tau_{sf} + \tau(\tau_{fs}-1)\tau_{sf} + \tau_{sf}(-1+\tau_{fs}-\tau_{sf}\tau_{fs})]\big] \tag{14}$$

$$\tau_0 = -\frac{(1-\tau_{fs})(R_s + \tau_{sf}R_{fc}^{ss}) + (\tau_{sf}-\tau)(\tau R_{fc}^{ss} + \tau_{fs}R_s)}{2\pi^2 R_{fc}^{ss}(\tau_{sf}-\tau)(\tau_{sf}\tau_{fs}-\tau)}. \tag{15}$$

The tricritical point reads

$$R_s^{tc} = \frac{4\pi^2\big[-\tau^3 + \tau^2\tau_{sf} + \tau(\tau_{sf} - 2\tau_{sf}\tau_{fs}) + [1 + \tau_{fs}(-1+\tau_{sf})]\tau_{sf}\big]}{(1+\tau)(-1+\tau+\tau_{fs}-\tau_{sf})} \tag{16}$$

Thus for $R_s < R_s^{tc}$, g_3 is positive and the bifurcation is supercritical and stable. Otherwise the bifurcation is subcritical and unstable. The existence of this unstable branch indicates that a hysteretic loop could be traced by increasing and decreasing ϵ around zero [7].

In Ref. 2, the τ_{ij}'s were found by fitting the theoretical frequency to the experimentally determined frequency. The values found are $\tau = 0.58$, $\tau_{sf} = 0.7$ and $\tau_{fs} = 0.35$ which correspond to NaCl and glycerol used as fast- and slow-diffusing solutes respectively. With Eqs. 16 and 6 the separation between the CTP and the TCP can be obtained in terms of cross diffusion constants, namely,

$$R_s^{c2} - R_s^{tc} = 4\pi^2\frac{(\tau-\tau_{sf})(\tau_{sf}\tau_{fs}-\tau)}{(1+\tau)(-1+\tau-\tau_{sf}+\tau_{fs})}. \tag{17}$$

5. Hysteresis loop in the stationary branch

Since $g_3 < 0$ for $R_s > R_s^{tc}$, there is a hysteresis loop which could be traced by varying R_f around R_{fc}^{ss} of the stationary branch. As R_f is increased, the system will remain in the conductive state as long as $R_f < R_{fc}^{ss}$. When $R_f > R_{fc}^{ss}$ convective motion will begin. If R_f is then decreased, convective flow will stop at the turning point $R_f^{tp} < R_{fc}^{ss}$ (or equivalently at a certain $\epsilon_{tp} < 0$) An approximate value of this turning point can be found by adding a quintic term to the amplitude equation (9), so that we will have $dA/dt = \epsilon A - g_3 A|A|^2 - g_5 A|A|^4$. In order to compute g_5, we have to go to fifth order in the perturbation scheme of last section. After a tedious calculation we obtain that g_5 at the TCP reads

$$g_5 = \frac{3(1+\tau)(-1+\tau+\tau_{fs}-\tau_{sf})}{640(\tau-\tau_{sf})(\tau-\tau_{fs}\tau_{sf})\big(-1+\tau_{fs}^2\tau_{sf} + \tau_{fs}(1+\tau+\tau^2-2\tau_{sf}-\tau\tau_{sf})\big)} \tag{18}$$

In the limit $\tau_{sf}, \tau_{fs} \to 0$, (18) agrees with the the quintic term computed in Ref. 4 without cross diffusion, as it should be.
Considering the steady state of the fifth order amplitud equation, one is left with

$$\epsilon = g_3 \mathcal{A}^2 + g_5 \mathcal{A}^4 \tag{19}$$

From $\partial\epsilon/\partial\mathcal{A} = 0$, one obtains the amplitude of the turning point

$$(\mathcal{A}_{tp})^2 = \frac{-g_3}{2g_5}, \tag{20}$$

which is the minimum value of the stable branch

$$\mathcal{A}^2 = -\frac{g_3}{2g_5} + \frac{1}{2}\sqrt{\left[\frac{g_3}{g_5}\right]^2 + \frac{4\epsilon}{g_5}}, \tag{21}$$

from which the system jumps to the conductive state at ϵ_{tp} which can be calculated by using $\mathcal{A}_{tp}$ in the Eqn. (19), one finds

$$\epsilon_{tp} = -\frac{g_3^2}{4g_5}. \tag{22}$$

Since $g_3 < 0$ for $R_s > R_s^{tc}$, Eqn. (20) indicates that g_5 should be positive as well. In the limit τ_{sf}, $\tau_{fs} \to 0$, it is evident that $g_5 = 3(1 - \tau^2)/640\tau^2$ is positive. It is known that τ_{fs} and τ_{sf} are smaller than τ, a close inspection of (18) indicates that g_5 is indeed positive. Given the values of τ_{ij}, one could compute g_3 and g_5, and find the values of the hysteresis loop parameters ϵ_{tp} and $\mathcal{A}_{tp}$ given by (22) and (20) respectively. These parameters provide a rough estimate of the amount of hysteresis expected in an experiment.

Figure 1 summarizes the main parameters of the hysteresis loop which we have just computed.

6. Conclusions

The location of the TCP for the stationary branch and the CTP had already been found with [6] and without [4] cross diffusion. It has been pointed out that since these points are well separated in parameter space, a study of the dynamics in their vicinity should be experimentally accessible. In this paper we computed a quintic coefficient of the amplitude equation in terms of cross diffusion constants which had been neglected so far. The quintic term allows us to roughly determine the expected amount of hysteresis in the vicinity of the TCP. It also allows us to predict the crossover from critical behavior $(A^2 \sim \epsilon)$ to tricritical behavior $(A^2 \sim \epsilon^{1/2})$ with $\epsilon = (R_f - R_{fc}^{ss})/R_{fc}^{ss}$, a prediction that should be feasible to test experimentally.

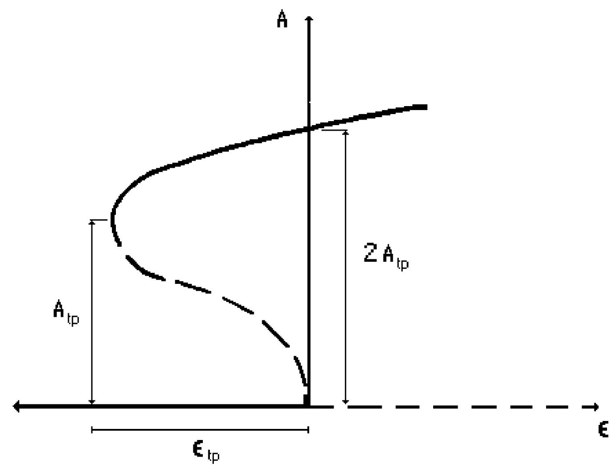

FIGURE 1. Hysteresis loop diagram. As R_f approaches R_{fc}^{ss} (or equivalently, as a negative ϵ approaches zero) the system will remain in the stable conductive state. When $R_f > R_{fc}^{ss}$ (or ϵ becomes positive) the system will jump to the convective state with amplitude $2\mathcal{A}_{tp}$. If R_f is then decreased, convection will stop at the turning point ϵ_{tp} when it has an amplitude $\mathcal{A}_{tp}$, not at $\epsilon = 0$. In the figure, solid lines represent stable states, whereas dashed lines represent unstable states.

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