Rayleigh-Marangoni convection in a critical fluid: A tale of two crossovers

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We show that if we take a thin layer of fluid where surface tension effects are supposed to dominate and gradually bring the mean temperature of the layer closer and closer to the liquid vapor critical point, then first there is a crossover from Marangoni to Rayleigh-Benard convection and thence to a convection whose onset is determined by the Schwarzchild criterion.

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I. INTRODUCTION

A recent experiment [1] on the convective instability in a pure fluid near its second order critical point has clearly revealed the crossover of the temperature difference required for the onset of convection from a Rayleigh criterion [2] dominated regime to a Schwarzchild criterion [3] dominated regime. This crossover is brought about as one approaches (i.e., the mean temperature of the convection cell approaches) the critical point leading to an enormous increase in compressibility. The Rayleigh criterion corresponds to an incompressible fluid while the Schwarzchild one corresponds to a compressible fluid. In the Rayleigh picture, the buoyancy force causes the hot fluid to rise—an effect which is opposed by the viscous drag. In the resulting dynamics if the hot fluid loses its heat due to thermal diffusion faster than it can rise, then convection cannot occur. This picture leads to the formation of a dimensionless variable called the Rayleigh number defined by

$$R=\frac{\alpha(\Delta T)gd^3}{\lambda\nu}, \tag{1.1}$$

where α is the thermal expansion coefficient, ΔT is the temperature difference between the bottom and top layers, g is the acceleration due to gravity, d is the depth of the liquid, λ is the thermal diffusivity, and ν is the kinematic viscosity. Convection occurs if R is greater than some critical value R_0 and thus the critical temperature difference ΔT_c for the onset of convection is

$$\Delta T_c=\frac{R_0\lambda\nu}{\alpha g d^3}. \tag{1.2}$$

If the fluid is near its second order phase transition point, then the static properties as well as the dynamic properties are strongly affected by the critical fluctuations. The strong fluctuations near the critical point are characterized by a correlation length ζ, which diverges (i.e., becomes infinitely big) as one approaches the critical temperature T_c. For a temperature very close to T_c, the behavior of ζ is scale invariant and can be written as [4]

$$\zeta=\zeta_0\left(\frac{T-T_c}{T_c}\right)^{-\mu}=\zeta_0 t^{-\mu}, \tag{1.3}$$

where μ is a critical exponent which is about 0.63 for the pure fluid. The thermal expansion coefficient diverges strongly as T approaches T_c and one has

$$\alpha\simeq\zeta^2 \tag{1.4}$$

for large ζ. The heat transport coefficient λ (the thermal diffusivity) shows critical slowing down and for large ζ,

$$\lambda\simeq\zeta^{-1}. \tag{1.5}$$

The viscous coefficient has a very weak divergence and we will ignore that over here without any significant error. Using Eqs. (1.3)–(1.5) in Eq. (1.2), we see that

$$\Delta T_c\simeq\zeta^{-3} \tag{1.6}$$

on the basis of the Rayleigh criterion and hence as the mean temperature of the cell approaches the critical point, the temperature gradient for onset of convection should approach zero.

However, for the extremely compressible fluid, the stability criterion involves the finite density difference due to an infinitesimal pressure difference. If a parcel of hot fluid is given an upward displacement ''d,'' then due to the temperature difference δT with the surrounding at this new position, it will see a favorable density difference

$$\frac{\delta\rho}{\rho}=\alpha\,\delta T. \tag{1.7}$$

The stabilizing density gradient would be provided by the pressure difference which is $\delta P=\rho g d$ and leads to a density difference

$$\frac{\delta\rho}{\rho}=\chi_T\delta P=\chi_T\rho g d. \tag{1.8}$$

From Eqs. (1.7) and (1.8), the onset of convection occurs if $\Delta T>\Delta T_c^{(s)}$ given by

$$\Delta T_c^{(s)}=\frac{\chi_T\rho g d}{\alpha}. \tag{1.9}$$

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Now, as $\zeta \to \infty$, $\chi_T \sim \zeta^2$, and $\alpha \sim \zeta^2$, so that ΔT_c acquires a finite value as opposed to the zero obtained from Eq. (1.6). The criterion shown in Eq. (1.9) is referred to as the Schwarzchild criterion. For a pure fluid the crossover from Eq. (1.6) to Eq. (1.9) has been beautifully demonstrated in the experiment of Kogan, Murphy, and Meyer [1].

We now consider the other mechanism [5,6] for the onset of convection—the effect of surface tension. If on the free surface there is a fluctuation causing a variation in temperature across the surface, then the surface tension σ which is a function of temperature is no longer constant across the surface and leads to an unbalanced force $(\partial\sigma/\partial x)\delta x$ per unit length. This force can be estimated from $(\partial\sigma/\partial T)\Delta T d$ and is analogous to the buoyancy force $\alpha\rho\Delta T g d^3$. The dimensionless number corresponding to the Rayleigh number of Eq. (1.1) is now

$$M = \frac{\left|\frac{\partial\sigma}{\partial T}\right|(\Delta T)d}{\rho\nu\lambda} \tag{1.10}$$

and is known as Marangoni number. Convection sets in as M becomes equal to a critical value M_0 and the critical (ΔT) is given by

$$\Delta\tilde{T}_c = \frac{M_0\rho\nu\lambda}{d\left|\frac{\partial\sigma}{\partial T}\right|}. \tag{1.11}$$

The relevance of the Rayleigh mechanism and the Marangoni mechanism can be judged from a comparison of Eqs. (1.2) and (1.11). In a given situation we can estimate the critical temperature difference required to see a buoyancy driven convection by looking at Eq. (1.2) and the temperature difference required to see a surface tension driven convection by using Eq. (1.11). The mechanism which yields a smaller value of the critical ΔT is the dominating mechanism. Clearly if d is large ΔT_c will be smaller and thermal expansion will dominate and if d is small, $\Delta\tilde{T}_c$ will be smaller and surface tension will dominate. The crossover thickness d_c is found from

$$d_c^2 = \frac{R_0}{M_0}\frac{\left|\frac{\partial\sigma}{\partial T}\right|}{\alpha\rho g}, \tag{1.12}$$

for $d \gg d_c$, the Rayleigh criterion holds and for $d \ll d_c$, it is pure Marangoni.

The criterion in Eq. (1.11) is obtained on the basis of the incompressibility assumption. Now, if we approach the critical point, then the surface tension vanishes according to [4]

$$\sigma \sim \zeta^{-2} \tag{1.13}$$

which means $\partial\sigma/\partial T \sim \zeta^{-2+1/\mu}$ and consequently

$$\Delta T_c \sim \zeta^{1-1/\mu} \tag{1.14}$$

since $1/\mu > 1$, $\Delta T_c \to 0$ as the critical point is approached. If we now consider the fluid to be compressible, which we must as it approaches the critical point, then the effect of compressibility will show up and instead of $\Delta\tilde{T}_c \to 0$ it will eventually saturate at $\Delta\tilde{T}_c^{(s)}$ given by Eq. (1.9).

We now imagine starting a convection experiment with $d \ll d_c$ and the mean temperature away from the critical temperature T_c. The onset of convection will be surface tension dominated. We now let the mean temperature approach T_c. From Eq. (1.12), we see that

$$d_c \sim \zeta^{-2+1/2\mu} \tag{1.15}$$

which implies that d_c decreases as we approach T_c. For $d_c = d$, there will be a crossover from Marangoni to Rayleigh behavior, the temperature corresponding to this crossover is given by the correlation length ζ_c, such that

$$d_c = d. \tag{1.16}$$

For $\zeta > \zeta_c$, the Rayleigh criterion will dominate and eventually for $\zeta \gg \zeta_c$, we will have a crossover to the Schwarzchild effect. Thus there will be two crossovers of this kind of an experiment from Marangoni to Rayleigh followed by another from Rayleigh to Schwarzchild. In the two subsequent sections, we will use the equations of hydrodynamics to establish the above result. In Sec. II, we provide a detailed derivation of the governing equations. This is necessary because the two previous approaches to Rayleigh convection in a compressible fluid led to equations which appeared to be very different from each other, although they seemed to yield critical Rayleigh numbers pretty close to each other. We provide a careful derivation in which if the surface fluctuations are dropped the previous results on Rayleigh-Benard convection appear with the connection between the two prior approaches apparent. In Sec. III, we solve the system of equations to formally arrive at the crossover described above.

II. MATHEMATICAL MODEL

In this section, we will set up the required equations of linear stability analysis from which the condition for destabilization of the conduction state will be obtained. The two relevant equations are the Navier Stokes equation for the velocity field $\vec{v}$, and the heat diffusion equation. The Navier Stokes equation reads (in presence of gravity)

$$\frac{\partial\vec{v}}{\partial t} + (\vec{v}\cdot\vec{\nabla})\vec{v} = -\frac{\vec{\nabla}P}{\rho} + \nu\nabla^2\vec{v} + g\hat{z}, \tag{2.1}$$

where P is the pressure and g the acceleration due to gravity. The heat diffusion equation reads

$$T\left(\frac{\partial}{\partial t}\delta Q + (\vec{v}\cdot\vec{\nabla})\delta Q\right) = \tilde{\lambda}\nabla^2\delta T, \tag{2.2}$$

where δQ is the entropy fluctuation and δT is the temperature fluctuation. These two relations need to be supplemented by the equation of continuity which reads

$$\frac{\partial \rho}{\partial t}+\vec{\nabla}\cdot(\rho\vec{v})=0. \tag{2.3}$$

The steady conduction state solution corresponds to all $\partial/\partial t=0, \vec{v}=0, \partial P/\partial z=-\rho g, \rho=\rho_0(z)$ and a linear temperature profile $T(z)=T_1+[(T_2-T_1)/d]z$, where T_1 and T_2 are the temperatures of the lower and upper plates, respectively, and d is the thickness of the fluid layer. To test the stability of the conduction state, we consider the fluctuations $\delta\vec{v}$, δP, $\delta\rho$, and δT of the velocity, pressure, density, and temperature fields and linearize the equations of motion [Eq. (2.1)–(2.3)] in terms of these variables.

We first need to determine the steady state density $\rho_0(z)$. To do so, we note that a variation in ρ with z is caused by variation of temperature and pressure. Consequently,

$$\frac{\partial \rho_0}{\partial z}=\frac{\partial \rho}{\partial P}\frac{\partial P}{\partial z}+\frac{\partial \rho}{\partial T}\frac{\partial T}{\partial z}=\frac{\rho_0}{d}(-A_1+A_2), \tag{2.4}$$

where

$$A_1=\rho_0 g\chi d \quad \text{and} \quad A_2=\alpha(\Delta T). \tag{2.5}$$

Linearization of Eq. (2.3) about the conduction state with $\rho=\rho_0(z)$ and $\vec{v}=0$ yields

$$\frac{\partial}{\partial t}\delta\rho+w\frac{\partial \rho_0}{\partial z}+\rho_0(\vec{\nabla}\cdot\delta\vec{v})=0. \tag{2.6}$$

At this point, we will introduce a simplification—we will be studying the stationary instability of the conduction state which implies that we will be interested in the critical value of R and M, at which the time dependence of the fluctuations vanish. So in Eq. (2.5) and in all subsequent equations, we will set $\partial/\partial t=0$. With this specification, Eq. (2.6) becomes

$$\rho_0(\vec{\nabla}\cdot\delta\vec{v})=-(\delta w)\frac{\partial \rho_0}{\partial z}. \tag{2.7}$$

We now turn to Eq. (2.1) and linearizing about the conduction state

$$-\rho_0\nu\nabla^2\delta v_i=-\left(\frac{\partial}{\partial x_i}+\frac{A_1}{d}\delta_{i3}\right)\delta P+\alpha g\rho\,\delta T\,\delta_{i3}. \tag{2.8}$$

Returning to Eq. (2.8), taking a divergence, operating with $(\partial/\partial z+A/d)$ and appropriately scaling variables to make them dimensionless, we get

$$\nabla^2\nabla^2 w+A_2\nabla^2\left(\frac{\partial w}{\partial z}\right)+A_1(A_2-A_1)\nabla^2 w=R\nabla_1^2\theta, \tag{2.9}$$

where ∇_1^2 is the Laplacian in the x-y plane. This equation is identical to Eq. (4.8) of Gitterman obtained by a different set of manipulations.

We now turn to the entropy equation and write the entropy fluctuation as

$$T\,\delta S=C_P\left(\delta T-\frac{\alpha V T}{C_P}\delta P\right) \tag{2.10}$$

Linearizing Eq. (2.2) about the conduction state, keeping $\partial/\partial t=0$, and carrying out the rescalings we have

$$\nabla^2\theta=-w(1-A), \tag{2.11}$$

where

$$A=\frac{\alpha T d g}{C_P\Delta T}=\frac{A_1}{A_2}\left(1-\frac{C_V}{C_P}\right). \tag{2.12}$$

Our Eq. (2.11) agrees with Eq. (1.10) of Gitterman and Steinberg.

We now examine the numerical values of the coefficients A_1 and A_2. For relative temperatures $t\sim10^{-4}$, it is clear from such an examination that A_1 and A_2 are numerically small but the ratio A_1/A_2 is close to unity for a ΔT which is of the order of a micro kelvin. At $t\sim10^{-4}$, $C_V\ll C_P$ and the factor A of Eq. (2.12) is consequently close to unity. The net result is that A_2 and $A_1(A_2-A_1)$ can be dropped in Eq. (2.9), but A needs to be retained in Eq. (2.11). It should be noted that in such experiments T_c can never be reached because there is a finite heat current and so t will never really become significantly smaller than 10^{-4}. In such a situation, Eqs. (2.9) and (2.11) reduce to

$$\nabla^4 w=-R\nabla_1^2\theta, \tag{2.13}$$

$$\nabla^2\theta=-w[1-A], \tag{2.14}$$

the system arrived at by Carles and Ugurtas [7]. To see the equivalence of Eqs. (14)–(19) of Carles and Ugurtas and our Eqs. (2.13) and (2.14), we note that Eqs. (15)–(17) of Ref. [7] can be written for the stationary state as

$$\nabla^2\vec{v}_1=\vec{\nabla}P_1+\rho_1\hat{z} \tag{2.15}$$

while Eq. (14) reads $\vec{\nabla}\cdot\vec{v}_1=0$. Taking a divergence of the former leads to $\nabla^2 P_1=-\partial\rho_1/\partial z$ or $\nabla^2(\partial P_1/\partial z)=-\partial^2\rho_1/\partial z^2$. If we now operate the z direction velocity profile with ∇^2,

$$\nabla^2\nabla^2 w_1=\frac{\partial}{\partial z}\nabla^2 P_1+\nabla^2\rho_1=\nabla_1^2\rho_1=-\frac{F}{a}\nabla_1^2 T_1, \tag{2.16}$$

where $\nabla_1^2=\partial^2/\partial x^2+\partial^2/\partial y^2$. A proper rescaling of w_1 and T_1 makes the above equation identical to Eq. (2.13) above. The identical rescaling of Eq. (18) of Ref. [7] now leads to Eq. (2.14) in the stationary limit by straightforward algebra. Since our Eqs. (2.13) and (2.14) were obtained from a technique similar to Gitterman and Steinberg and eventually, keeping the leading terms, we arrive at a system identical to that of Carles and Ugurtas, we believe that the two approaches give the same result. Thus we have provided a derivation which shows clearly the connection between the two different forms existing in the literature—that due to Gitterman [3] and that due to Carles and Ugurtas [7]. We now discuss the boundary conditions [10], where we will have to introduce the effect of the surface tension. At the lower plate (taken to be conducting), the ''no-slip'' condition implies that

$$w=\frac{\partial w}{\partial z}=\theta=0 \quad \text{at} \quad z=0. \tag{2.17}$$

The top surface is free and if from the mean position of $z=1$ there is a fluctuation η, then

$$w=\frac{\partial \eta}{\partial t} \quad \text{at} \quad z=1. \tag{2.18}$$

For a stationary instability $\partial\eta/\partial t=0$ and hence

$$w=0 \quad \text{at} \quad z=1. \tag{2.19}$$

If the interface is very weakly conducting, then we can approximate it as insulating and then

$$\frac{\partial \theta}{\partial z}=0 \quad \text{at} \quad z=1. \tag{2.20}$$

Now for the force balance on the interface, the stress tensor is

$$T_{ij}=-P\delta_{ij}+\rho\nu\left(\frac{\partial v_i}{\partial x_j}+\frac{\partial v_j}{\partial x_i}\right). \tag{2.21}$$

The change in the normal component of the stress is $2\sigma\cdot$curvature, while the horizontal component of the stress tensor has to be provided by the gradient of the surface tension. The surface being characterized by the deflection $\eta(x,y)$, we have the unit vectors (ignoring quadratic powers of η) given by: normal $\hat{n}=(-\partial\eta/\partial x,-\partial\eta/\partial y,1)$ and the tangential $\hat{t}=(1,0,\partial\eta/\partial x)$. The normal force balance on the surface gives [10]

$$\tilde{C}\left(\frac{\partial^2}{\partial z^2}+3\nabla_1^2\right)\frac{\partial w}{\partial z}+(A_2-A_1)\nabla^2 w=(\nabla_1^4-\tilde{B}\nabla_1^2)\eta, \tag{2.22}$$

where $\tilde{C}=\rho\nu\lambda/2\sigma d$ is the crispation number and $\tilde{B}=\rho g d^2/2\sigma$ is the Bond number. From the tangential stress balance we get

$$\frac{\partial^2 w}{\partial x^2}-\frac{\partial^2 T}{\partial z^2}-(A_2-A_1)\frac{\partial w}{\partial z}+M\frac{\partial^2(\theta+\eta)}{\partial x^2}=0 \quad \text{on } z=1. \tag{2.23}$$

We now choose the coordinate system such that the axis of the rolls coincide with the y axis and thus there is no y dependence in w and θ. The x dependence is periodic with wave number a in dimensionless units and the z-dependent functions for w and θ are $F(z)$ and $G(z)$, respectively, in such a way that $w=F(z)e^{iax}$ and $\theta=G(z)e^{iax}$. So in the final analysis, $F(z)$ and $G(z)$ satisfy the following equations for Rayleigh-Marangoni convection in a compressible fluid [under the approximation that $A_1, A_2 \ll 1$ and $A\sim O(1)$]

$$(D^2-a^2)^2F=Ra^2G,$$
$$(D^2-a^2)G=-(1-A)F \tag{2.24}$$

with

$$F=DF=G=0 \quad \text{on} \quad z=0,$$
$$F=DG=0 \quad \text{on} \quad z=1,$$
$$\tilde{C}(D^2-3a^2)DF=a^2(\tilde{B}+a^2)\eta \quad \text{on} \quad z=1,$$
$$(D^2+a^2)F+Ma^2(a+\eta)=0 \quad \text{on} \quad z=1. \tag{2.25}$$

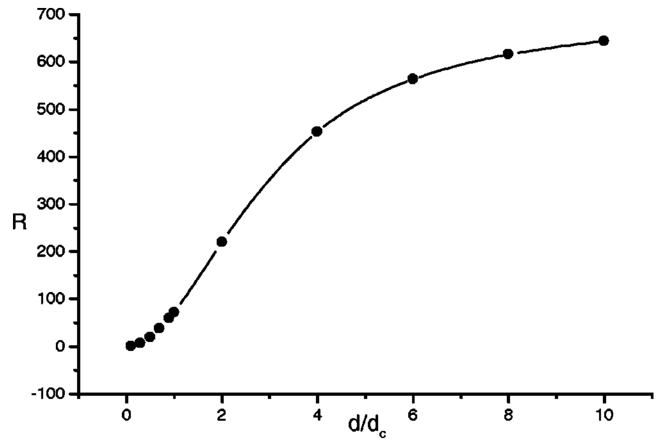

FIG. 1. Plot of R_c versus (d/d_c).

In the next section, we will analyze the solution of Eqs. (2.24) under the boundary condition of Eq. (2.25) to arrive at the relation between R and M that is necessary for the solvability. To end this section, we note that we are defining (as has been conventional in the literature) M in terms of ΔT, where $\Delta T/d$ is the temperature gradient in the layer.

An inconsistency in this approach was recently pointed by Rabin [8], but as shown in the experiment of Schatz *et al.* [9], the conventional definition gives a very account of the experiment and hence we will follow the conventional definition of the Marangoni number.

III. ANALYSIS

We begin with the observation that the surface fluctuations η will be determined by the ratio $\tilde{C}/\tilde{B}=\nu\eta/gd^3$. Close to the critical point, the shear viscosity and hence ν has a weak divergence while the thermal diffusivity vanishes as the inverse of the correlation length ζ, which makes $\tilde{C}/\tilde{B}$ small near the critical point. Consequently in the following analysis, we will ignore the effect of the surface fluctuations. We will use a variational function $G(z)$ for the temperature fluctuations, which is known to be very accurate [9] for the pure Marangoni convection. Our technique will be to (i) use a trial function for the temperature variable $G(z)$; (ii) solve for the velocity profile from the velocity equation and ensure that all boundary conditions are satisfied; (iii) now satisfy the temperature equations in the mean to find the relation between R and M.

Accordingly, we choose

$$G(z)=z\left(1-\frac{z}{2}\right), \tag{3.1}$$

where an overall prefactor has been set equal to be unity, in anticipation of the fact that in the homogeneous system that we have, it cannot eventually matter. The function $G(z)$ incorporates the conducting walls at $z=0$ and the insulating walls at $z=1$. The solution of $(D^2-a^2)^2F=Ra^2G$, now reads

$$F(z)=A_1\cosh az+A_2\sinh az+A_3z\cosh az+A_4z\sinh az+\frac{R}{a^2}\left(-\frac{z^2}{2}+z-\frac{2}{a^2}\right), \tag{3.2}$$

where A_1, A_2, A_3, and A_4 are constants which have to be obtained from the boundary conditions. Satisfying the temperature profile equation in the mean, we arrive at the final result:

$$\Delta T=\frac{R_0\lambda\nu}{\alpha g d^3}\left(\frac{1}{1+\frac{R_0}{M_0}\left(\frac{d_c}{d}\right)^2}\right)+\frac{\alpha T g d}{C_P}, \tag{3.3}$$

where d_c is the crossover thickness defined in Eq. (1.12), and

$$R_0(a)=\frac{2a^4(CS-a)\left(1+\frac{a^2}{3}\right)}{CS\left(\frac{2a^2}{3}-6\right)-\frac{3}{2}aS^2+\frac{13C^2}{a}+\frac{1}{a}(a^2-4)(aS+4C)-\frac{7}{6}a^3+a+\frac{3}{a}} \tag{3.4}$$

and

$$M_0(a)=\frac{4(CS-a)\left(1+\frac{a^2}{3}\right)}{(S-a)^2}, \tag{3.5}$$

where $C=\cosh a$ and $S=\sinh a$.

If we work with a thickness d which is much smaller than d_c when one is well away from the critical temperature, then as expected

$$\Delta T\simeq\frac{M_0\lambda\nu}{\alpha g d d_c^2}+\frac{\alpha T g d}{C_P}\simeq\frac{M_0\lambda\nu}{\alpha g d d_c^2} \tag{3.6}$$

which is the pure Marangoni convection. As the mean temperature T of the convection cell is lowered, d_c decreases and ΔT tends towards a value closer to that for pure Rayleigh convection. It should be noted that even if the critical phenomena is unimportant and the compressibility effect exhibited by the second term on the right hand side of Eq. (3.3) is absent, Eq. (3.3) provides a clean analytic answer to the problem of studying the crossover from Marangoni to Rayleigh convection as d is increased. For this purpose, it is best to exhibit the result in a plot of R_c vs d/d_c, where R_c is the usual critical Rayleigh number which for this problem is around 700 with $a=a_c\simeq 2.0$. From Eq. (3.3), we obtain in this limit

$$R=\frac{R_0M_0\left(\frac{d}{d_C}\right)^2}{M_0\left(\frac{d}{d_C}\right)^2+R_0} \tag{3.7}$$

which is a function of the wave number a, and has to be minimized with respect to a to give R_c. The plot in Fig. 1 shows that even when $d=10d_c$, the effect of surface tension is to cause a reduction of 8% in the pure buoyancy driven Rayleigh number.

Returning to our present problem, we interpret the changing temperature as a changing d_c for a fixed d, and hence as we go closer to the critical point we move from a small value of d/d_c to a later much larger value of d/d_c as the critical point is approached.

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