Onset of convection in a binary mixture near the plait point

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(Received 13 October 1999)

Recent experiments on the onset of convection near the liquid-vapor critical point of ^{3}He have shown the crossover from the Rayleigh criterion to the Schwarzschild criterion for the threshold of convection as the critical point is approached. In contrast we show that for ^{3}He-^{4}He mixtures near the liquid-vapor critical point (plait point), the Rayleigh criterion would hold right through. Interestingly enough, this is a consequence of the proper boundary conditions. This prediction should be easy to test experimentally.

PACS number(s): 44.25.+f, 47.27.Te, 64.70.Fx

The onset of Rayleigh Benard convection in a single component fluid is very well understood [1]. The Rayleigh criterion for the onset can be understood qualitatively, the critical Rayleigh number calculated quantitatively, and the result successfully compared with experiments. If the convecting fluid is very close to the critical point and hence extremely compressible, the Rayleigh criterion has to be changed to the Schwarzchild criterion [2]. The quantitative calculation, first done by Steinberg and Gitterman [3,4] has lately been given a much simpler reformulation by Carles and Ugurtas [5]. The results show a smooth crossover from the Rayleigh criterion to the Schwarzschild criterion as the fluid is brought closer and closer to the critical point. Very recently a clean experimental verification of this result has been given by Kogan, Murphy, and Meyer [6], who worked with ^{3}He very near its critical point. The single most important feature is that under the Rayleigh criterion, the critical temperature difference ΔT_c for the onset of convection would go to zero as the critical point is approached, whereas the crossover from Rayleigh to Schwarzschild implies that there is a finite ΔT_c even at the critical point. This finite value of ΔT_c at extreme criticality, has indeed been observed by Kogan, Murphy, and Meyer. Denoting the distance from critical temperature by $\epsilon[\epsilon=(T-T_c)/T_c]$, Kogan, Murphy, and Meyer observe that ΔT_c decreases with ϵ for $\epsilon>10^{-2}$, but then levels off at a value which is completely consistent with the Schwarzschild criterion.

A strong qualitative twist to the Rayleigh convection in a single component fluid can be obtained if we consider a binary fluid. The important additional features are the possibility of having a density gradient due to the concentration gradient and the fact that a mass current can be produced by a temperature gradient (Soret effect) and vice versa (Dufour effect). It is customary to write the mass current $\vec{j}$ as

$$\vec{j}=-D\left(\vec{\nabla}c+\frac{k_T}{T_m}\vec{\nabla}T\right), \tag{1}$$

where D is the mass diffusion, k_T is the thermodiffusion which describes the efficiency of a temperature gradient in producing a mass current, and T_m is a mean temperature. The quantities which classify the nature of the convective instability are the separation ratio ψ, and the Lewis number S defined as

$$\psi=-\frac{\beta}{\alpha_{P,c}}\frac{k_T}{T},\quad S=\frac{D}{D_T}, \tag{2}$$

$$\beta=-\frac{1}{\rho}\left(\frac{\partial\rho}{\partial c}\right)_{P,T} \quad\text{and}\quad \alpha_{P,c}=-\frac{1}{\rho}\left(\frac{\partial\rho}{\partial T}\right)_{P,c}, \tag{3}$$

with c as the concentration of one of the fluids in the mixture, and $D_T=\lambda/C_{P,c}$ with λ the thermal conductivity at zero mass current, and $C_{P,c}$ the specific heat at constant pressure and concentration. If a mixture with positive k_T is heated from below, an upward concentration gradient would be established and for positive β this would mean a decreasing density as one went upwards and a stable situation would ensure. Thus, in the mixture with negative ψ, the convective instability does not occur when heated from below. Instead, to see the usual Rayleigh instability for negative ψ, the fluid mixture needs to be heated from above. When heated from below, the mixture undergoes a Hopf bifurcation and above a critical Rayleigh number R_0, oscillatory convection sets in [7,8]. To see the usual Rayleigh convection, when heated from below, one needs a positive value of ψ. For large positive ψ/L, the onset occurs as long wavelength rolls (zero wave number). The instability which occurs when heated from above also occurs as long wavelength rolls. The oscillatory instability which occurs for $\psi>0$, when heated from below, occurs as finite wavelength rolls. Various features of this convection have been experimentally observed by Lee, Lucas, and Tyler [9], Bloodworth *et al.* [10], Kolodner *et al.* [11], and Moses and Steinberg [12].

In this paper, we ask the question, what would happen if the mixture were extremely compressible? This happens near the plait point which is the critical point for the liquid-vapor transition of the mixture. We consider mixtures with positive ψ for which stationary convection occurs when heated from below. The behavior of the static responses of such mixtures was investigated by Griffiths and Wheeler [13] and the dynamic responses by Mistura [14] with crossover effects being studied by Luettmer-Strathmann and Sengers [15] and Folk and Moser [16]. The consequence of the above investigations is that the separation parameter diverges as $\zeta^{1-\alpha/\nu}$ as the

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critical point is approached where ζ is the correlation length. Our prediction is that very close to the critical point, the usual Rayleigh Benard stationary convection will occur when the mixture is heated from below and that this onset will be in the form of long wavelength rolls with the Schwarzschild criterion never coming into play. These results are qualitatively different from the behavior near the ordinary critical point discussed above. The static and dynamic critical properties of the mixture ^{3}He-^{4}He have been studied near the plait point by Cohen, Dingus, and Meyer [17,18]. Consequently we believe that this mixture will serve as the ideal candidate for the experimental verification of the above contentions.

To establish the above result, we need to set up the relevant hydrodynamic equations for the mixture near the plait point. The field variables are velocity $\vec{v}(\vec{r},t)$, the local temperature $T(\vec{r},t)$, the density $\rho(\vec{r},t)$, and $c(\vec{r},t)$, the concentration of one of the species [for the ^{3}He-^{4}He mixture, we will consider $c(\vec{r},t)$ as the concentration of ^{4}He]. The density satisfies the continuity equation

$$\frac{\partial \rho}{\partial t}+\vec{\nabla}\cdot(\rho\vec{v})=0. \tag{4}$$

The velocity satisfies the Navier-Stokes equation

$$\frac{\partial \vec{v}}{\partial t}+(\vec{v}\cdot\vec{\nabla})\vec{v}=-\frac{\vec{\nabla}P}{\rho}+\nu\nabla^2\vec{v}+\vec{g}. \tag{5}$$

The concentration satisfies the conservation law

$$\frac{\partial c}{\partial t}+(\vec{v}\cdot\vec{\nabla})c=-\vec{\nabla}\cdot\vec{j}=D\nabla^2\left(c+\frac{k_T}{T_m}T\right). \tag{6}$$

The heat conduction equation in terms of the temperature variable is most easily obtained by starting with the form of entropy flow in Landau and Lifshitz and using temperature, pressure, and concentration as the independent variables. Standard manipulations lead to

$$C_{P,c}\left[\frac{\partial T}{\partial t}+(\vec{v}\cdot\vec{\nabla})T\right]-T\left(\frac{\partial v}{\partial T}\right)_{P,c}\left[\frac{\partial P}{\partial t}+(\vec{v}\cdot\vec{\nabla})P\right]-k_T\left(\frac{\partial \mu}{\partial c}\right)_{P,T}\left[\frac{\partial c}{\partial t}+(\vec{v}\cdot\vec{\nabla})c\right]=\kappa\nabla^2 T \tag{7}$$

(κ is the conductivity at vanishing concentration gradient) and on using Eq. (6),

$$C_{P,c}\left[\frac{\partial T}{\partial t}+(\vec{v}\cdot\vec{\nabla})T\right]-T\left(\frac{\partial v}{\partial T}\right)_{P,c}\left[\frac{\partial P}{\partial t}+(\vec{v}\cdot\vec{\nabla})P\right]=\lambda\nabla^2 T+\frac{Dk_T^2}{T_m}\left(\frac{\partial \mu}{\partial c}\right)_{P,T}\nabla^2 c, \tag{8}$$

which together with Eqs. (4)–(6) constitute the set of relevant hydrodynamic equations.

The conduction state in a large aspect ratio fluid layer of height L in the z direction is characterized by

$$\vec{v}=\vec{0},\ \frac{\partial P}{\partial z}=-\rho g,\quad T=T_1-\frac{\Delta T}{L}z,$$

$$\vec{j}=\vec{0},\quad \Delta c=-\frac{k_T}{T_m}\Delta T. \tag{9}$$

In the above, T_1 is the temperature of the bottom plate and $\Delta T=T_1-T_2$ where T_2 is the temperature of the upper plate. It is the stability of the above solution of Eqs. (4)–(6) and (8) against the formation of convection rolls that needs to be studied. To this end, we carry out a linear stability analysis in the perturbations $\delta\vec{v}$ (components u, v, and w), δP, $\delta\rho$, $\delta T, \delta c$. It is convenient to use the constitutive relation

$$\frac{\delta\rho}{\rho}=\frac{1}{\rho}\left(\frac{\partial\rho}{\partial P}\right)_{T,c}\delta P-\alpha_{P,c}\delta T-\beta\delta c \tag{10}$$

to eliminate the density fluctuation.

At this stage, a great simplification was noticed by Carles and Ugurtas. Linearizing Eq. (4) in the perturbations leads to

$$\frac{\partial}{\partial t}\delta\rho+\rho_0(\vec{\nabla}\cdot\delta\vec{v})+w\frac{\partial\rho_0}{\partial z}=0. \tag{11}$$

For a stationary instability, this implies $\vec{\nabla}\cdot\delta\vec{v}=-w(\partial/\partial z)ln\,\rho_0$. Now the variation of ρ_0 across the cell height L is practically negligible for all reasonable values of L and this implies that the perturbative velocity $\delta\vec{v}$ is approximately divergence free. Using this constraint, we can write down the linearized set of equations in a fairly straightforward manner. Keeping in mind the boundary conditions that need to be applied, it is convenient to use instead of δc, fluctuation $\delta\sigma$ defined as $\delta\sigma=\delta c+(k_T/T_m)\delta T$. The gradient of $\delta\sigma$ gives the current. All variables are dimensionless: we scale time by $\lambda/L, \delta T$ by $\Delta T, \delta\sigma$ by Δc and all distances by L. This leads to

$$\nabla^4 w=-(1+\psi)R\nabla_1^2\theta-\psi R\nabla_1^2\eta,$$

$$\nabla^2\eta=-\frac{w}{S},$$

$$(1-\mu^2 S)\nabla^2\theta=-w(1-A)+\mu^2 S\nabla^2\eta, \tag{12}$$

where θ and η are the scaled forms of δT and $\delta\sigma$, $\mu^2=(k_T^2/TC_{P,c})(\partial\mu/\partial c)_{P,T}$, $A=\alpha_{P,c}LTg/C_{P,c}\Delta T$ and R is the Rayleigh number defined as

$$R=\frac{\alpha_{P,c}\Delta TL^3 g}{\nu\frac{\lambda}{C_{P,c}}}. \tag{13}$$

The derivative ∇_1^2 stands for $(\partial^2/\partial x^2)+(\partial^2/\partial y^2)$. For formation of convection rolls, the solution will be periodic in the x and y directions with wave number $\vec{a}$ having the components a_1 and a_2. Convecting solutions for the various fields will have the form $[f_i(z)e^{i(a_1x+a_2y)}]$. Writing $D=d/dz$, the above equations become

$$(D^2-a^2)^2 f_1=(1+\psi)Ra^2 f_2+\psi Ra^2 f_3,$$

$$(D^2-a^2)f_3=-\frac{f_1}{S},$$

$$(1-\mu^2 S)(D^2-a^2)f_2=-f_1(1-A)+\mu^2 S(D^2-a^2)f_3. \tag{14}$$

The boundary conditions that need to be imposed at $z=0$ and $z=1$ are $f_1=Df_1=0$, $f_2=0$ (bounding plates of high thermal conductivity) and $Df_3=0$ (bounding surfaces impenetrable). Using trial solutions in the spirit of Gutkowicz-Krusin, we find

$$R=\frac{\dfrac{(\pi^2+a^2)^3}{a^2}\dfrac{1-\mu^2 S}{1+\mu^2-A}}{\left[\left(1+\psi+\dfrac{\psi}{S}\dfrac{1-\mu^2 S}{1+\mu^2-A}\right)G_1-\dfrac{\psi}{S}\dfrac{1-\mu^2 S}{1+\mu^2-A}G_2\right]}, \tag{15}$$

where

$$G_1=1-\frac{16a\pi^2\cosh^2 a/2}{(\pi^2+a^2)^2(a+\sinh a)}, \tag{16}$$

and

$$G_2=\frac{\pi^2}{a^2}\left[1+\frac{(\pi^2-3a^2)}{(\pi^2+a^2)}\frac{\coth a/2}{a}-\frac{1+\cosh a}{a+\sinh a}\coth a/2\right]. \tag{17}$$

From Eq. (15), the critical temperature difference ΔT for the onset of convection with wave number a is

$$\Delta T=\frac{\nu\lambda}{C_{P,c}}\frac{1}{\alpha_{P,c}L^3 g}\frac{(\pi^2+a^2)^3}{a^2}\times\frac{(1-\mu^2 S)}{\left[1+\mu^2+\psi\left(1+\dfrac{1}{S}\right)\right]G_1-\dfrac{\psi}{S}(1-\mu^2 S)G_2}+\frac{\alpha_{P,c}LTg}{C_{P,c}}\frac{(1+\psi)G_1}{\left[1+\mu^2+\psi\left(1+\dfrac{1}{S}\right)\right]G_1-\dfrac{\psi}{S}(1-\mu^2 S)G_2}, \tag{18}$$

the principal result of this paper. To find the actual ΔT_c we need to minimize the above expression as a function of a.

Since, the above expression is to be considered near the plait point, we need to examine the critical behavior of the various thermodynamic and transport properties involved. From Griffiths and Wheeler, we know that $C_{P,c}$ and $\alpha_{P,c}$ are proportional to $\zeta^{\alpha/\nu}$ and that $(\partial c/\partial\mu)_{P,T}\sim\zeta^{\gamma/\nu}$. Combining these results with the transport coefficients found by Mistura, $\mu^2\sim\zeta^{-\alpha/\nu}$, $S\sim\zeta^{-1+\alpha/\nu}$, $\psi\sim\zeta^{1-\alpha/\nu}$, $\nu\sim\zeta^{X_\eta/\nu}$ and $\lambda\sim\zeta^0$. Sufficiently close to criticality when ζ is large, Eq. (18) simplifies to

$$\Delta T=\frac{S}{\psi}\frac{\nu\lambda}{C_{P,c}}\frac{1}{\alpha_{P,c}L^3 g}\frac{(\pi^2+a^2)^3}{a^2}\frac{1}{G_1-G_2}+\frac{\alpha_{P,c}LTg}{C_{P,c}}\frac{SG_1}{G_1-G_2}. \tag{19}$$

It is the second term on the right hand side which is present only when the system is near the plait point and the compressibility is taken into account. For the single component fluid it leads to the Schwarzschild criterion. If we examine the functions G_1 and G_2, then we find that G_1 varies between $1-8/\pi^2$ and 1 as a goes from zero to infinity, while G_2 is $-(24-2\pi^2/3a^2)$ for $a<<1$ and increases to π^2/a^2 for $a>>1$. Thus, G_1 is always $+ve$ and G_2 is $-ve$ for all reasonable values of a and the combination G_1-G_2 is always $+ve$. The first term in ΔT is consequently positive for all negative ψ and the second term is always positive. Now $S/\psi<<S$ close to T_c and hence the second term might appear to dominate, which would be the Schwarzschild effect. Indeed, for idealized stress-free boundaries this would be the case. But for the realistic boundaries considered here, the second term is $O(a^2)$ for $a<<1$ and finite for $a>>1$. The first term as a function of a^2 is finite as $a^2\to 0$ and diverges as a^4 for $a>>1$.

Consequently, the critical Rayleigh number is obtained for $a\to 0$ and is given by

$$R=\frac{S}{\psi}\frac{3\pi^6}{24-2\pi^2}\simeq 677\frac{S}{\psi}. \tag{20}$$

Having noted the irrelevance of the compressibility from the above variational calculation, we can mention that the exact result corresponding to Eq. (20) is $R=720S/\psi$.

In closing, we want to reemphasize the importance of boundary conditions in obtaining the above result. To do this, let us return to Eq. (18) and without making any assumption regarding the critical behavior of the parameters involved study the right hand side as a function of a. For $a\to 0$, the first term on the right hand side without any approximation is $S/\psi[3\pi^6/(24-2\pi^2)+Ba^2]$, where B is a positive constant. The second term is exactly $(\alpha LTg/C_{pc})[(1+\psi)S/\psi(1-\mu^2 S)][1-(8/\pi^2)](3a^2/24-2\pi^2)$. The $O(a^2)$ correction to the leading term of the right hand side of Eq. (18) is consequently always positive forcing $a=0$ to be a minimum. The simplification that we have exhibited in Eq. (19) by taking the asymptotic critical behavior into account is only to exhibit the potentially important Schwarzschild term. It is well known that in a real experiment, there would be crossover effects which would mean the passage from Eq. (18) to Eq. (19) is going to be complicated as a function of reduced temperature and concentration. However, the boundary condition on the mass current makes this irrelevant for our final answer. Due to the boundary condition, the Schwarzschild term is never important. We can recast the result in another fashion: in a single component fluid like ^{3}He-^{4}He near its liquid-vapor critical point, the compressibility effect changes the hydrodynamic effect and a crossover from a Rayleigh criterion to the Schwarzschild criterion occurs. In a mixture like ^{3}He-^{4}He near its liquid-vapor critical point, the zero mass current boundary condition of hydrodynamics is so strong that it completely masks the critical effect and keeps the hydrodynamics unaltered at the Rayleigh criterion. It should be noted that the boundary condition which makes this work is the zero mass current condition at the bounding surfaces — a fact which will be true for all experimental cells. What can differ, is the thermal boundary on the plates and this will change the prefactor of S/ψ in Eq. (20), e.g., for thermally insulating boundaries the prefactor is 120 instead of 677. We end with the claim that for a convection experiment done on a liquid mixture (heated from above for negative ψ and heated from below for positive ψ), the Schwarzschild effect will not be seen and instead the usual Rayleigh criterion of Eq. (20) will hold. The only requirement for this is that the mixture shows a finite k_T which for dynamical purposes identify it as a binary mixture. Since k_T has a strong divergence, this is not a problem for any of the critical fluids.

One of the authors (K.S.D.) would like to thank CSIR India for partial financial assistance.

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