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Phenomenological Phase Transition Approach to the Two-Component Rayleigh–Bénard Problem: Tricritical Behavior

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A previously introduced Landau "potential" for the description of the basic features of the two-component Rayleigh–Bénard problem is shown to be endowed with the relevant properties of the classical Landau singular free energy used in the study of equilibrium tricritical phenomena. Predictions of relevance to experiments on convective instability are also given.

KEY WORDS: Instability; convection; bifurcation; tricritical.

1. LANDAU POTENTIAL FOR THE TWO-COMPONENT RAYLEIGH–BÉNARD PROBLEM

With the use of a Landau "potential" a rather simplified and compact description of the basic features of thermohydrodynamic instability and *natural convection* can be achieved.$^{(1-4)}$ Moreover, it turns out that on quantitative grounds all theoretical predictions agree quite satisfactorily with the presently available experimental data.[3] The justification for such an agreement lies in the fact that in Bénard convection, as in some other convective instabilities, correlation lengths are of true macroscopic size, e.g., they take the dimension of the sample and not merely a few thousand

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[3] For a recent account see Bergé.$^{(5)}$

angstroms. Thus, side boundary effects prevent one from seeing *nonclassical* corrections.[4] On the other hand, corrections to the Landau phenomenological description simply improve the results in extremely small regions or lead to negligible corrections, at least in the simplest Bénard problem.(3,6)

In the present note we consider the model problem of a horizontal two-component liquid layer heated from below and discuss the behavior to be expected in the neighborhood of a "tricritical" point.(2) This designation has been introduced following ideas from equilibrium statistical mechanics[5] and it denotes a point where *three* coexisting "phases" become identical. Although the tricritical point in the two-component Bénard problem may very well not be easily accessible to experiment,[6] the present study yields useful light about the behavior to be expected every time that lines of subcritical instability (hard-mode, first-order transition) end at points of exchange of stabilities (soft-mode, continuous transition). This is also the case for homeotropic nematic liquid layers heated from below(9),[7] or every time that a non-Boussinesquian fluid property is taken into consideration.(3)

Following the notation introduced in an earlier paper,(2) the model problem under consideration leads to the study of the following Landau "potential"[8]

$$\Phi = \frac{32}{3}\pi^4 r^2\left[\frac{27\pi^4}{4} - R\left(S + 1 + \frac{S}{r}\right)\right]V^2 + \frac{4}{9}\left[\frac{27\pi^4}{4}(1 + r^2) - R\right]V^4 + \frac{V^6}{6} \qquad (1)$$

which here corresponds to the classical Landau free energy used in equilibrium phase transitions. The following notation has been introduced: R is

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[4] For a comprehensive theoretical background see Graham.(6)

[5] Our work follows in the spirit of ideas developed for metamagnets, helium mixtures, and the laser. See Refs. 7. General definitions and notation can be found in the pioneering work of Griffiths; see, for instance, Ref. 8.

[6] Actually this will not be the case with experiments using macromolecular solutions, or in the nematic case discussed below where the parameters can be varied at will.

[7] For experimental results and a truncated mode approach see Ref. 10.

[8] The full Navier–Stokes, Fick–Soret, and Fourier equations cannot be derived from a *true* potential with an *unrestricted* variational principle behind them. It is only a truncated version of these equations, as discussed in Refs. 2 and 4, that can be made to derive from a potential. An irrelevant constant term has been omitted in (1). When non-Boussinesquian properties or any other supplementary symmetry-breaking mechanisms are incorporated the potential takes account of them with odd powers in the order parameter. The case discussed here corresponds to a homogeneous approximation in the case of stress-free, permeable, and heat-conducting boundaries. None of the approximations is conceptually important. Inhomogeneities, i.e., spatial variations of the convective (ordered) state, are incorporated by generating a Landau–Ginzburg functional, as discussed in Refs. 1, 4, and 5.

the Rayleigh number (a dimensionless measure of the thermal gradient across the layer), S is the Soret separation parameter (a dimensionless measure of the strength of Soret separation due to the thermal gradient), and r is the Lewis number (a dimensionless ratio between the mass and heat diffusivities which is of order 10^{-2} in standard liquid mixtures). V is the unknown and plays the role of order parameter in the problem (it corresponds to the amplitude of the relevant convective velocity field).

For later convenience we write (1) in the following manner:

$$\Phi = \tfrac{1}{2}A(R,S,r)V^2 + \tfrac{1}{4}B(R,S,r)V^4 + \tfrac{1}{6}C(R,S,r)V^6 \tag{2}$$

where

$$A(R,S,r) = (64/3)\pi^4 r^2\big[(27/4)\pi^4 - R(S + 1 + S/r)\big] \tag{3}$$

$$B(R,S,r) = (8/9)\big[(27/4)\pi^4(1 + r^2) - R\big] \tag{4}$$

$$C(R,S,r) = 1 \tag{5}$$

The Lewis number can be taken as a parameter. For a given value r the coordinates of the "tricritical point" are obtained by setting $A = B = 0$. We have

$$R^* = (27/4)\pi^4(1 + r^2), \qquad S^* = -r^3/(1 + r^2)(1 + r)$$

Notice that R^* is positive, whereas S^* is negative, in accordance with results of (linear) stability analysis.$^{(11)}$ On the other hand, this and all other properties to be discussed below are not specific to the truncation imposed to generate the potential (1). Higher order truncations lead to the same picture.[9]

The crucial, albeit standard assumption now is to consider the quantities A, B, and C as analytical functions around the "tricritical point." Then, a straightforward consequence of the scheme is the classical behavior of the order parameter

$$V \sim (R - R^*)^{1/4} \quad \text{at} \quad S = S^* \quad \text{and} \quad R > R^* \tag{6a}$$

$$V \sim |S^* - S|^{1/4} \quad \text{at} \quad R = R^* \quad \text{and} \quad S < S^* \tag{6b}$$

Obviously, the relevant "response functions" (specific heat-like variable, "susceptibilities," etc.) will exhibit classical behavior.

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[9] A comprehensive discussion of various truncations of the thermohydrodynamic equations and the numerical results found is given in Ref. 12. In this paper there is a wealth of information concerning hysteretic phenomena and metastability.

2. TRICRITICAL BEHAVIOR IN BINARY MIXTURES HEATED FROM BELOW

Comparison with experiment cannot be made yet, as the experimental work on either binary mixtures(13) or homeotropic nematic layers(14) is still underway. Computer simulation of the dynamics of binary mixtures is also in progress in our department, but no valid result is available at the moment. Thus merely for the record we shall list here some of the results predicted by our theory, focusing in detail only on the behavior of the susceptibility associated with the order parameter and the phase separation picture.

We introduce the *conjugate* quantities to R and S,

$$X = -(\partial\Phi/\partial R)_{S,r} \tag{7a}$$

$$Y = -(\partial\Phi/\partial S)_{R,r} \tag{8a}$$

Thus we have

$$X = (32/3)\pi^4 r^2 V^2/(1 + r^2) + 2V^4/9 \tag{7b}$$

$$Y = 144\pi^8 r(1 + r)(1 + r^2)V^2 \tag{8b}$$

The expected behavior around the tricritical point is

$$X \sim (R - R^*)^{1/2} \text{ along } S = S^*, \text{ and } X \sim |S^* - S|^{1/2} \text{ at } R = R^* \tag{9}$$

Y follows the same power law.

A specific heat-like variable is

$$c = R(\partial X/\partial R)_{S,r} = -R(\partial^2\Phi/\partial R^2)_{S,r} \tag{10}$$

From Eq. (1) we get

$$c = \frac{1024\pi^8 r^4 R}{9(1 + r^2)}\left[\frac{64\pi^4 r^2(R - R^*)}{3(1 + r^2)} + 144\pi^8 r(1 + r)(1 + r^2)(S - S^*)\right]^{-1/2} \tag{11}$$

which shows the expected $(-1/2)$ singularity in $(R - R^*)$ or $(S^* - S)$. Another "response" function is $(\partial Y/\partial S)_R$. We have

$$\left(\frac{\partial Y}{\partial S}\right)_R = -\frac{144^2}{2}\pi^{16}r^2(1 + r^2)^2(1 + r)^2 \times\left[\frac{64}{3}\,\frac{\pi^4 r^2(R - R^*)}{r^2 - 1} + 144\pi^8 r(1 + r)(1 + r^2)(S^* - S)\right]^{-1/2} \tag{12}$$

For this variable the following behavior is predicted:

$$(\partial Y/\partial S)_R \sim (R - R^*)^{-1/2} \quad \text{along} \quad S = S^* \tag{13a}$$

$$(\partial Y/\partial S)_R \sim |S^* - S|^{-1/2} \quad \text{along} \quad R = R^* \tag{13b}$$

Yet another "response" function is $(\partial Y/\partial R)_S$, whose singular behavior at the tricritical point is

$$(\partial Y/\partial R)_S \sim (R - R^*)^{-1/2} \quad \text{along} \quad S = S^* \tag{14a}$$

$$(\partial Y/\partial R)_S \sim |S^* - S|^{-1/2} \quad \text{along} \quad R = R^* \tag{14b}$$

For the "susceptibility" associated with the order parameter we merely consider that **h** is some external field, e.g., a preimposed flow along the horizontal, a preimposed pressure gradient in the vertical, a *true* external field (magnetic, electric) in a nematic layer or in a mixture response to it, etc. Then we have $\chi_{R,S} = (\partial V/\partial h)_{R,S,r}$ together with $(\partial \Phi/\partial V) = h$. From the use of this definition and the expression (1) or (2) it follows that

$$\chi_{R,S}^{-1} = 5V^4 + (8/3)V^2\left[(27/4)\pi^4(1 + r^2) - R\right] + \left[(27/4)\pi^4 - R(S + 1 + S/r)\right](64/3)\pi^4 r^2 \tag{15}$$

This susceptibility-like quantity has in fact been measured in various ways for the case of Rayleigh–Bénard convection in single-component liquid layers(5,15) and it seems easy to introduce in experiments with binary mixtures or homeotropic liquid layers. The expected singularities at the tricritical point are

$$\chi_{R,S} \underset{h\to 0}{\sim} (R - R^*)^{-1} \quad \text{along} \quad S = S^* \tag{16a}$$

$$\chi_{R,S} \underset{h\to 0}{\sim} |S^* - S|^{-1} \quad \text{along} \quad R = R^* \tag{16b}$$

with coefficients readily obtainable from Eq. (15) or from the corresponding similar expression for different boundary conditions.

We have also studied the behavior expected along continuous (soft-mode, second-order) transitions and estimated the critical slowing down. The results can be found in Ref. 2 and 4 and nice agreement exists with the only available data.(16) We shall not dwell on this here. Let us now illustrate the "phase separation" picture provided by our potential (1).[10]

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[10] For those readers interested in currently fashionable problems, it is to be noted that the underlying equations that generate (1) constitute a natural extension of the Lorenz model to the two-component Bénard problem.(2,3) The first transition in the Lorenz model is second order, whereas here the two-component Lorenz model exhibits hard-mode excitations or continuous transitions depending on the parameters R and S. Exceptionally low values of the Lewis number eliminate the first-order transitions, as shown in Refs. 11 and 17.

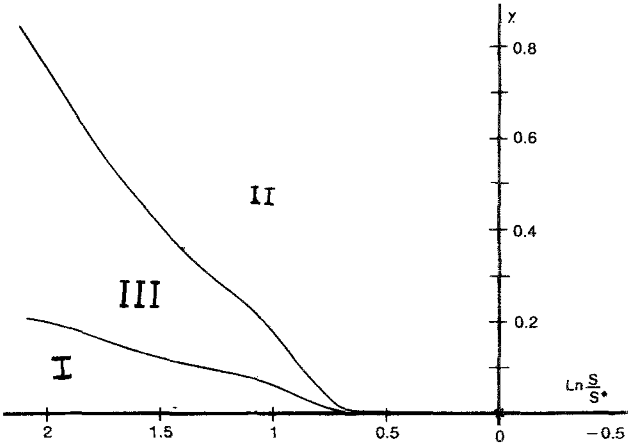

Fig. 1. "Phase separation" in the two-component Rayleigh–Bénard problem. The origin corresponds to the point where to its left no exchange of stabilities is expected. This indicates that transitions from the motionless state to a convective regime are expected to show hysteretic behavior, metastability, and overstable motions, which are the genuine properties of first-order (hard-mode) transitions, or subcritical instability as it is called in the fluid mechanics jargon. From the origin to its right a line of second-order (continuous, soft-mode) transitions develops. Along this line there is exchange of stabilities (critical slowing down). The figure corresponds to the value $r = 0.01$ (Lewis number for standard liquid mixtures). Thus S^* gives, together with R^*, the location of a "tricritical point." Region I corresponds to the motionless state, region II is the convecting state, and in region III there is coexistence of motion with rest. The two heavy lines separating these regions are lines of first-order transition according to nonlinear analysis.

Phase separation can be illustrated by plotting the variable Y, Eq. (8), in front of the Soret separation parameter, S. Using Eq. (8b), we have

$$Y = 144\pi^8 r(1 + r)(1 + r^2) \times \left[\frac{64\pi^4 r^2(R - R^*)}{3(1 + r^2)} + 144\pi^8 r(1 + r)(1 + r^2)(S^* - S) \right]^{1/2} \tag{17}$$

This is the relationship shown in Fig. 1 for the Lewis number $r = 0.01$. By varying the Lewis number, a three-dimensional picture is readily obtained. In Fig. 1, two lines of first-order transition separate three regions: a disordered phase (the motionless region), an ordered (convective) phase, and the region of coexistence of motion and rest. These two lines of hard-mode excitations end at the origins, at which, for $S > S^*$, the continuous transition line develops.

Recently, the study presented here has been extended to the case of time-dependent (limit cycle) states of the fluid layer.(18)

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