

PRETRANSITIONAL EFFECTS IN THE RAYLEIGH-BENARD INSTABILITY

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I. INTRODUCTION

Recently there has been a growing interest in pretransitional effects in hydrodynamic regime transitions². Particular emphasis is put on the interesting resemblance that these pretransitional effects bear to those occurring in equilibrium phase transitions. One of the most striking analogies is the amplification of thermal fluctuations near the transition point.

The simplest example of a hydrodynamic instability is the Rayleigh-Bénard or convective instability which occurs in a horizontal fluid layer heated from below. When the temperature gradient reaches a certain critical value stationary convection sets on spontaneously. The fundamental physical process which causes this instability is the conversion of the energy released by the buoyancy force into the kinetic energy of the convective motion. Stationary convection sets on when the rate of energy transfer from the gravitational field to the convective motion balances the rate of viscous dissipation of energy of the convective motion.

Following essentially the stochastic treatment of Zaitsev and Shliomis³ we first analyze the hydrodynamic fluctuations in the pretransitional state. Next we discuss the analogies between pretransitional effects in second-order phase transitions and in hydrodynamic regime transitions. Finally we suggest the use of light scattering and Brownian motion to observe experimentally the anomalous fluctuations near the instability point.

II. HYDRODYNAMIC FLUCTUATIONS NEAR THE INSTABILITY POINT

The basic equations describing the convective instability are⁴

$$\frac{\partial}{\partial t} \nabla^2 w = \nu \nabla^4 w + \alpha g \left(\frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial y^2} \right), \quad (2.1)$$

$$\frac{\partial \theta}{\partial t} = \beta w + \chi \nabla^2 \theta \quad (2.2)$$

Here w is the vertical component of the velocity, θ is the perturbation in the temperature, $-\beta$ is the (vertical) temperature gradient and α , χ and ν are the thermal expansion

coefficient, thermal diffusivity and kinematic viscosity, respectively. For the sake of simplicity we consider a fluid layer with free boundaries, in which case \mathbf{w} and θ can be written as

$$[w(\vec{r}, t), \theta(\vec{r}, t)] = \sum_{n=1}^{\infty} \int d\vec{\kappa} [w(n, \vec{\kappa}, t), \theta(n, \vec{\kappa}, t)] \times \exp(i \vec{\kappa} \cdot \vec{x}) \sin(n\pi z/d). \quad (2.3)$$

Here $\vec{\kappa} = (\kappa_x, \kappa_y)$ is the horizontal wavevector, $\vec{x} = (x, y)$ is the horizontal position vector and d is the thickness of the fluid layer. Substituting Eq. (2.3) in Eqs. (2.1) and (2.2) one obtains the following equations for the Fourier amplitudes

$$\frac{\partial}{\partial t} w(n, \vec{\kappa}, t) = -\nu k^2 w(n, \vec{\kappa}, t) + \alpha g \kappa^2 k^{-2} \theta(n, \vec{\kappa}, t) + F^{(w)}(n, \vec{\kappa}, t), \quad (2.4)$$

$$\frac{\partial}{\partial t} \theta(n, \vec{\kappa}, t) = \beta w(n, \vec{\kappa}, t) - \chi k^2 \theta(n, \vec{\kappa}, t) + F^{(\theta)}(n, \vec{\kappa}, t), \quad (2.5)$$

where $k^2 = \kappa^2 + (n\pi/d)^2$. In order to study the hydrodynamic fluctuations we have introduced fluctuating forces in the hydrodynamic equations. As usual we assume that these fluctuating forces are gaussian markoffian δ -correlated random processes and for the fluctuation intensities of these forces we use the equilibrium expressions.

$$\begin{aligned} \langle F^{(\ell)}(n, \vec{\kappa}, t) F^{(\ell)}(n', \vec{\kappa}', t')^* \rangle &= \\ &= 2 Q^{(\ell)} \delta_{n, n'} \delta(\vec{\kappa} - \vec{\kappa}') \delta(t - t'), \\ &(\ell = w, \theta), \end{aligned}$$

$$Q^{(w)} = \frac{i}{2\pi^2 d} \frac{k_B T \kappa^2}{\rho k^2} \nu k^2, \quad (2.6)$$

$$Q^{(\theta)} = \frac{i}{2\pi^2 d} \frac{k_B T^2}{\rho C_p} \chi k^2$$

The instability sets in as soon as one of the eigenvalues of the two coupled linear equations (2.4) and (2.5) goes to zero, i.e. as soon as the determinant of the coefficient matrix becomes equal to zero

$$\nu \chi k^4 - \frac{\alpha \beta g \kappa^2}{k^2} = 0. \quad (2.7)$$

Introducing the Rayleigh number $R = \alpha \beta g d^4 / \nu \chi$, it follows that Eq. (2.7) is satisfied for $R = k^6 d^4 \kappa^{-2}$. The lowest value of R for which this is realized is $R_c = 27 \pi^4 / 4$, obtained for $n = 1$ and $\kappa = \kappa_c = \pi / \sqrt{2} d$.

The two coupled equations (2.4) and (2.5) have two eigenvalues for each wavevector $(\vec{\kappa}, n\pi/d)$ and correspondingly there are two eigenmodes. We shall only consider the eigenmodes corresponding to the smallest root and with wavevectors close to the critical one, i.e. with $n = 1$ and κ in the vicinity of κ_c . Indeed those are the modes that develop anomalous fluctuations near the instability point. For these modes we can write the equation

$$\frac{\partial}{\partial t} \psi(l, \vec{\kappa}, t) = -\lambda(l, \kappa) \psi(l, \vec{\kappa}, t) + F^{(\psi)}(l, \vec{\kappa}, t), \quad (2.8)$$

where $\lambda(l, \kappa)$ is the root of the coupled equations (2.4) and (2.5) that goes to zero for $R = R_c$ and $\kappa = \kappa_c$. The corres-

ponding mode is a linear combination of $w(l, \vec{\kappa}, t)$ and $\theta(l, \vec{\kappa}, t)$. For $R = R_c$ and $\kappa = \kappa_c$ one can write

$$\psi(l, \vec{\kappa}, t) = w(l, \vec{\kappa}, t) + \left(\frac{\alpha g \nu}{3\beta \chi}\right)^{1/2} \theta(l, \vec{\kappa}, t). \quad (2.9)$$

Similarly the intensity $Q^{(\psi)}$ of the random force $F^{(\psi)}(l, \vec{\kappa}, t)$ is a linear combination of $Q^{(w)}$ and $Q^{(\theta)}$. From Eq. (2.9) it follows that

$$\begin{aligned} Q^{(\psi)} &= Q^{(w)} + (\alpha g \nu / 3\beta \chi) Q^{(\theta)} \\ &= Q^{(w)} \left(1 + \alpha g T / c_p \beta\right) \\ &\approx Q^{(w)}, \end{aligned} \quad (2.10)$$

as for typical liquids $\alpha g T / c_p \beta \sim 10^{-6}$.

From (2.8) one obtains

$$\langle \psi(l, \vec{\kappa}, 0) \psi(l, \vec{\kappa}', t)^* \rangle = \frac{Q^{(\psi)}}{\lambda(l, \kappa)} e^{-\lambda(l, \kappa)t} \delta(\vec{\kappa} - \vec{\kappa}'). \quad (2.11)$$

This result clearly shows that for R close to R_c and for κ close to κ_c , i.e. where $\lambda(l, \kappa) \rightarrow 0$, the fluctuations in $\psi(l, \vec{\kappa}, t)$ decay very slowly (critical slowing down) and have a very large intensity ("critical opalescence"). Expressing $w(l, \vec{\kappa}, t)$ and $\theta(l, \vec{\kappa}, t)$ in terms of $\psi(l, \vec{\kappa}, t)$ one finds, close to the instability point,

$$\begin{aligned}
 \langle W(1, \vec{k}, 0) W(1, \vec{k}', t)^* \rangle &= \\
 &= \left(\frac{\chi}{\chi + \nu} \right)^2 \frac{Q^{(\psi)}}{\lambda(1, \kappa)} e^{-\lambda(1, \kappa)t} \delta(\vec{k} - \vec{k}'),
 \end{aligned}
 \tag{2.12}$$

$$\begin{aligned}
 \langle \theta(1, \vec{k}, 0) \theta(1, \vec{k}', t)^* \rangle &= \\
 &= \left(\frac{\nu}{\chi + \nu} \right) \frac{3\beta}{\alpha g} \frac{Q^{(\psi)}}{\lambda(1, \kappa)} e^{-\lambda(1, \kappa)t} \delta(\vec{k} - \vec{k}').
 \end{aligned}
 \tag{2.13}$$

III. ANALOGIES WITH PRETRANSITIONAL EFFECTS IN SECOND ORDER PHASE TRANSITIONS

In second-order phase transitions there are two well-known pretransitional effects : near the critical point the fluctuations in the order parameter become very large and they decay very slowly. These effects can be explained from the behavior of the susceptibility associated with the order parameter near the transition point. Starting from the Landau expression for the free energy

$$F = F_0 + \frac{1}{2} \int d\vec{r} \left[a \varepsilon \eta(\vec{r})^2 + b (\nabla \eta(\vec{r}))^2 \right],$$

where $\eta_{\vec{k}}$ is the order parameter and $\varepsilon = T/T_c - 1$ one obtains for the \vec{k} -th spatial Fourier component of the susceptibility

associated with η

$$\chi(\vec{k}) = \frac{1}{(2\pi)^3} \frac{1}{a\varepsilon + bk^2}, \quad (3.1)$$

which diverges for $T \rightarrow T_c$ and $k \rightarrow 0$. From the well-known connection between fluctuations and susceptibility

$$\begin{aligned} \langle |\eta(\vec{k})|^2 \rangle &= k_B T \chi(\vec{k}) \\ &= \frac{1}{(2\pi)^3} \frac{k_B T}{a\varepsilon + bk^2}, \end{aligned} \quad (3.2)$$

it follows that the fluctuations in the order parameter become very large for $T \rightarrow T_c$. Further according to irreversible thermodynamics the decay rate of small deviations from equilibrium can be written as

$$\lambda = \frac{L}{\chi},$$

where L is an Onsager kinetic coefficient and χ is the susceptibility associated with the considered variable. According to the conventional theory of critical slowing down, the kinetic coefficient associated with the order parameter behaves regularly near T_c and since χ becomes large near T_c , λ becomes small

$$\lambda = \frac{L}{\chi(\vec{k})} = (2\pi)^3 L (a\varepsilon + bk^2). \quad (3.3)$$

As seen in the previous section, near the convective instability point, the fluctuations in the eigenmodes that become unstable, are very large and decay very slowly. In view of the analogy with the order parameter behavior in a second-order phase

transition, we will call ψ the order parameter for the hydrodynamic regime transition. The analogy in the pretransitional behavior of η and ψ becomes even more explicit upon analyzing the behavior of $\lambda(l, \kappa)$. For R just below R_c and for κ close to κ_c one can write

$$\lambda(l, \kappa) = a' \epsilon + b' (\kappa - \kappa_c)^2, \quad (3.4)$$

where in this case $\epsilon = 1 - R/R_c$, and the expansion coefficients are $a' = [\chi\nu/(\chi+\nu)](\kappa_c^2 + \pi^2/d^2)$ and $b' = 4\chi\nu/(\chi+\nu)$. Substituting Eq. (3.4) in (2.11) and taking $t = 0$ one obtains

$$\langle |\psi(l, \vec{\kappa})|^2 \rangle = \frac{Q(\psi)}{a' \epsilon + b' (\kappa - \kappa_c)^2}. \quad (3.5)$$

Notice the striking analogy between Eqs. (3.2) and (3.5) i.e. for the order parameter fluctuations in a second-order phase transition and in a hydrodynamic regime transition, respectively.

IV. EXPERIMENTAL POSSIBILITIES OF OBSERVATION

In this section we briefly consider the possibilities to observe experimentally the anomalous pretransitional fluctuations near the convective instability. In view of the great success of light scattering studies of critical phenomena near second-order phase transitions and because hydrodynamic instabilities are triggered by thermal fluctuations which can be probed by light scattering, it seems logical to first consider this tool for the investigation of pretransitional effects near hydrodynamic instability points. Since this topic has been reviewed recently by one of us (J.P.B.)⁵ we shall restrict ourselves to a few comments.

The fluctuations in the order parameter $\psi(l, \vec{\kappa}, t)$ are only strongly amplified for κ close to $\kappa_c = \pi/\sqrt{2}d$. Since the thickness of the fluid layer in which the instability takes place is typically of the order of 0.1 - 1 cm it follows that in order to observe the large fluctuations in the order parameter one has to use scattering angles as small as 10^{-3} - 10^{-4} radian. This certainly represents a non trivial experimental problem and so far no experimental evidence for anomalous fluctuations near the convective instability point has been produced.⁶

Fluid fluctuations can also be probed through the Brownian motion of suspended particles. Using Faxén's theorem⁷ it can be shown that the diffusion coefficient of a spherical Brownian particle can be written⁸ as

$$D_j = \int_0^{\infty} dt \langle \bar{v}_j^s(0) \bar{v}_j^s(t) \rangle ; j = x, y, z, \quad (4.1)$$

where $\bar{v}_j^s(t)$ is the average of $v_j(\vec{r}, t)$, the fluctuating fluid velocity field, over the surface of the Brownian particle. Using Eq. (2.12) for the fluctuating fluid velocity field it can be shown that the diffusion coefficient in addition to its regular Stokes-Einstein part contains a critical part such that

$$D = D^{reg} + D^{crit} = \frac{k_B T}{6\pi\eta a} + \frac{k_B T}{24\sqrt{3}\eta d} \epsilon^{-3/2} \quad (4.2)$$

As can be seen from Eq. (4.2) the ratio of D^{crit} to D^{reg} is determined by $\epsilon^{-3/2} a/d$. Consider $a \sim 1 \mu m$ and $d \sim 1 mm$; then at $\epsilon = 10^{-2}$, D becomes about 50% larger than its regular value. Given that the instability occurs at a temperature difference of about $10^\circ C$ between the lower and upper boundary, values of ϵ down to 10^{-4} are in experimental

reach without great difficulty. Photon correlation spectroscopy is an appropriate method to probe the dynamics of Brownian motion and by this method, diffusion coefficients can be determined very accurately. Consequently there should be no problem to observe the increase in the diffusion coefficient. With sufficient experimental accuracy it might even be possible to extract the value of the critical exponent.

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