ANALYSIS OF THE APPROACH TO THE CONVECTIVE INSTABILITY POINT

J.P. Boon (1) and H.N.W. Lekkerkerker

Faculté des Sciences, Université Libre Bruxelles, 1050 Brussels and Fakulteit van de Wetenschappen, Vrije Universiteit Brussel, 1050 Brussels, Belgium

## ABSTRACT

A spectral analysis is presented of the fluctuations in a horizontal fluid layer subject to a downward directed temperature gradient, which, for a critical value, drives the system in a convective instability state. It is found that the external force resulting from the combination of the temperature gradient and the gravitation force gives rise to a coupling between the heat diffusion mode and a shear mode. As a result of this mode coupling the damping constant of the heat diffusion mode goes to zero when the temperature gradient increases towards its critical value, i.e. the heat diffusion mode behaves like a "soft mode". The implications of the mode coupling and of the ensuing softening of the heat diffusion mode on the light scattering spectrum are discussed.

### INTRODUCTION

In a horizontal fluid layer heated from below a stationary convection mode appears spontaneously for a critical value of the downward directed temperature gradient. This is one of the simplest examples of a hydrodynamic instability, (well known in classical physics as the Bénard problem (2)).

One of the prime objects of investigations in hydrodynamic stability has been the determination of the values of the state parameters at which the transition from the stability regime to the instability domain occurs. So far little attention has been given to the process of initiation of non-equilibrium instabilities (see however references 3 and 4). The reason for this may be the difficulty to observe pretransitional phenomena in classical hydrodynamic experiments. However, it appears that modern experimental methods allow the investigation of the pretransitional states. Indeed, since instabilities are triggered by thermal fluctuations, light scattering spectroscopy which probes these fluctuations, appears as anappropriate tool to investigate the pretransitional fluctuations (5,6).

The Bénard problem, because of its simplicity, was chosen here to investigate pretransitional phenomena. Using hydrodynamic fluctuation theory, we show how the spectral features of the light scattering spectrum are modified when the fluid departs from equilibrium towards the instability critical point. The spectral changes are found to be most important for the central line. An additional Lorentzian component arises because the entropy fluctuations, which in an equilibrium one-component fluid decay in a single heat diffusion mode, decay into two non-propagating modes in the presence of the external force. When the system approaches the instability critical point the damping of one of these modes goes to zero indicating a "soft mode" type of behavior which suggests an analogy between hydrodynamic instability phenomena and structural phase transitions (7).

## SPECTRAL ANALYSIS OF THE FLUCTUATIONS

As is well known, the spectral intensity distribution of the polarized component of scattered light is proportional to  $S_\varepsilon(\underline{k},\omega)$  the spectral density of the kth spatial Fourier component of the fluctuation  $\delta\varepsilon$  in the optical dielectric constant. Here  $\underline{k}$  is the change in wave vector and  $\omega$  the change in frequency upon scattering. The dielectric constant can be considered as a function of the thermodynamic state of the system, so that  $\delta\varepsilon$  can be expressed in terms of the fluctuations  $\alpha_{\underline{i}}$  in the thermodynamic state variables  $A_{\underline{i}}$ . Consequently

$$S_{\epsilon} (\underline{k}, \omega) = \sum_{i,j} (\frac{\partial \epsilon}{\partial A_{i}}) (\frac{\partial \epsilon}{\partial A_{j}}) S_{\alpha_{i}\alpha_{j}} (\underline{k}, \omega).$$
(1)

The spectral densities of the fluctuations in the thermodynamic state variables occuring on the right hand side of equation (1) will be calculated here by assuming that also in non-equilibrium systems do the fluctuations on the average decay according to the appropriate hydrodynamic equations, i.e. the linearized equations for the fluctuations in the steady state variables. In the Bénard problem the steady state can be described as follows: an adverse linear temperature gradient is maintained steadily; there is no convective motion; and the gravitational force is balanced by the pressure gradient. Labeling the steady state variables with the superscript s one has

$$\mathbf{T}^{\mathrm{S}}(z) = \mathbf{T}_{\mathrm{o}} - \beta z ; \underline{\mathbf{v}}^{\mathrm{S}} = 0 ; \partial_{\mathbf{z}} \mathbf{p}^{\mathrm{S}}(z) = -g \boldsymbol{\rho}^{\mathrm{S}}(z)$$
(2)

where the subscript o denotes the value of a variable at the reference position, which for the sake of convenience is taken at the lower boundary (see figure 1). For the purpose of obtaining the linearized equations for the fluctuations in the steady state variables the equation for  $p^S(z)$  and  $\rho^S(z)$  need not be solved explicitly as long as  $|p^S(z) - p_o| \ll p$  and  $|\rho^S(z) - \rho_o| \ll \rho_o$  which conditions are usually satisfied for normal fluids and for actual experimental conditions (layer tickness  $\sim 0.1 - 1 \text{ cm}$ ).

In the present case, we choose to write the thermodynamic equations for the following set of variables:

$$\delta s = s - s^{S}; \quad \delta p = p - p^{S}; \quad \varphi = \partial_{X} v_{X} + \partial_{y} v_{y}; \quad v_{Z}; \quad (\nabla x \underline{v})_{Z}.$$

The above description of the velocity field is a consequence of the symmetry of the system under consideration. It can be shown that  $\varphi$  and  $v_z$  have the same transformation properties as  $\delta s$  and  $\delta p$  under the symmetry operations of the system, whereas  $(\nabla x \underline{v})_z$  does not and will therefore not couple to  $\delta s$  and  $\delta p$ .



Figure 1. Representation of the geometry of a fluid layer subject to an adverse temperature gradient.

One then obtains the following set of coupled linearized equations

$$(\partial_{t} - D_{T}\nabla^{2}) \delta s - (\alpha/\rho) D_{T}\nabla^{2} \delta p - (Cp/T) \beta v_{z} = 0,$$

$$(\partial_{t} - (\gamma-1) D_{T}\nabla^{2}) \delta p - (\rho/\alpha) (\gamma-1) D_{T}\nabla^{2} \delta^{S}$$

$$+ \phi/\chi_{s} - (\alpha\beta - \partial_{z}) v_{z}/\chi_{s} = 0,$$

$$(\partial_{t} - (\nu+\nu') \nabla^{2} + \nu' \partial_{z}^{2}) \phi - \nu' (\nabla^{2} - \partial_{z}^{2}) \partial_{z} v_{z}$$

$$+ \rho^{-1} (\nabla^{2} - \partial_{z}^{2}) \delta p = 0,$$

$$(\partial_{t} - \nu \nabla^{2} - \nu' \partial_{z}^{2}) v_{z} - \nu' \partial_{z} \phi + (g\chi_{s} + \rho^{-1} \partial_{z}) \delta p$$

$$- (g\alpha T/C_{p}) \delta s = 0.$$

$$(4)$$

Here is  $\alpha$  the thermal expansitivity,  $\chi_S$  the adiabatic compressibility,  $\gamma$  the ratio of the specific heats at constant pressure (C<sub>p</sub>) and at constant volume (C<sub>v</sub>), D<sub>T</sub> the thermal diffusivity,  $\nu$  the kinematic viscosity and  $\nu' = (\xi + \eta/3)/\rho$  where  $\eta$  is the shear viscosity and  $\xi$  the bulk viscosity.

As the system is supposed to be infinite in the x and y directions (i.e. the actual geometry of the system is such that the thickness of the fluid layer is much smaller than its dimensions in the x, y plane) a spatial Fourier transformation can be performed in these coordinates. For the sake of mathematical simplicity we consider hypothetical boundary conditions at the lower and upper plates such that the variables  $\delta s$ ,  $\delta p$ ,  $\phi$  and vz can also be Fourier transformed in the z direction. Further the set of equations (4) is Laplace transformed in the time variable. Considering the terms due to the presence of the temperature gradient and of the gravitation force as small, one can treat the dispersion equation of the Fourier-Laplace transformed coupled equations (4) by perturbation theory. To zeroth order one retrieves the usual modes for the system at equilibrium. From the higher order terms it is found that the non-propagating modes are weakly affected by it. Therefore the latter can safely be neglected here and one then obtains for the damping factors of the non-propagating modes

$$s_{\pm} = -\frac{k^{2}}{2} (\nu + D_{T}) \pm \frac{k^{2}}{2} [(\nu - D_{T})^{2} + 4\nu D_{T} \frac{R}{R_{c}(\underline{k})}]^{\frac{1}{2}}, \quad (5)$$

with

$$R/R_{c}(\underline{k}) = \frac{\alpha\beta g}{\nu D_{T}} \frac{k^{2} - k_{z}^{2}}{k^{6}} .$$
(6)

Here R is the Rayleigh number and  $R_c(\underline{k})$  its critical value for the mode with wave vector  $\underline{k}$ . Indeed, note that  $R/R_c(\underline{k})$  depends strongly on the wavelength of the fluctuation under consideration. This fact has important consequences for probing pretransitional phenomena with light scattering spectroscopy and will be discussed in the next section. For most liquids  $\nu > D_T$  and it is easily seen from equation (5) that in that case the external force induces a coupling such that the heat diffusion mode (S\_) behaves like a "soft mode" simultaneously with the broadening of the shear mode (S\_). When considering only the non-propagating modes, the set of equations (4) reduces to two coupled equations. These can easily be solved for  $\delta s(\underline{k},s)$  (the Fourier-Laplace transform of  $\delta s$ ), from which the appropriate correlation function is constructed to yield the spectral distribution of scattered light. One obtains

$$I_{\underline{k},\omega}^{\text{central}} \propto \left(\frac{\partial \varepsilon}{\partial s}\right)_{p}^{2} \langle |\delta s(\underline{k})|^{2} \rangle \langle A_{+} \frac{|s_{+}|}{s_{+}^{2} + \omega^{2}} + A_{-} \frac{|s_{-}|}{s_{-}^{2} + \omega^{2}}, \qquad (7)$$

with

$$A_{+} = 1 - A_{-} = \frac{(\nu k^{2} - s_{+})^{2}}{(\nu k^{2} - s_{+})^{2} + \nu D_{T} k^{4} R / R_{c}(\underline{k})}$$

One thus observes that the central peak for a system subject to an adverse temperature gradient consists of two Lorentzians (Note that in the limit  $R \rightarrow 0$ , i.e.  $\beta \rightarrow 0$  one retrieves the single heat diffusion line as expected for a fluid at equilibrium). The spectrum as given by equation (7) is illustrated in figure 2.

#### DISCUSSION

The most interesting result is undoubtedly the fact that the damping factor of one of the non-propagating modes goes to zero when the instability critical point is approached. From the expression of  $R/R_{\rm c}(\underline{k})$  given by equation (6), it is clear that for a given temperature gradient the effect of the external force will be most important for small values of k. This means that the most dramatic spectral changes are to be observed at small scattering angles. Indeed, the wavelength of the mode which is the first to become unstable is of the order of the vertical dimension of the system.



Figure 2. Representation of the central spectral components of a fluid subject to an adverse temperature gradient. The arrows indicate the modification of the width of the spectral components when the temperature gradient increases towards its critical value.

Presently available techniques used in light scattering spectroscopy should allow to probe modes which are expected to be affected in a fluid subject to an external force.

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