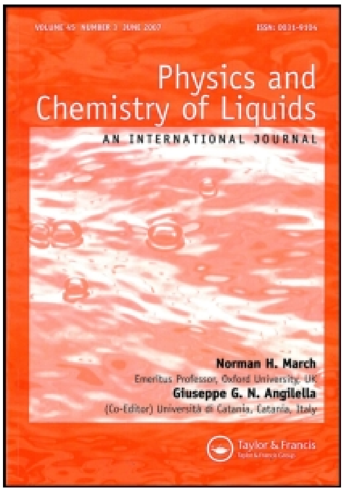

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Spectral Analysis of a Fluid under Thermal Constraint

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Abstract—A spectral analysis is presented to investigate the onset of the convective instability in a fluid subject to a linear temperature gradient. The hydrodynamic theory is developed for the case of a binary system, where the concentration of one of the components is small. Therefore the present results will be applicable to the case of a Brownian system. We consider exclusively those modes which correspond to the central components of the spectral distribution, i.e., the diffusion mode and the thermal diffusivity mode. One finds that those modes are effected by the presence of the external temperature gradient in such a way that the spectrum of the scattered light should exhibit an important narrowing of the thermal diffusivity peak and a slight narrowing of the diffusion peak when approaching the convective instability critical point. Only the thermal diffusivity mode is affected in the limit of a pure fluid.

1. Introduction

Instabilities occur in a wide range of situations in many different fields, including not only hydrodynamics and thermodynamics, but also chemical physics, biology, geophysics, urban growth, meteorology, and astrophysics. To date most of the work on these phenomena(1,2) has been devoted to macroscopic analyses and in particular to the study of the so-called marginal states. However, very little is known concerning the departure from the stability regime and about the dynamics governing the onset of the marginal state.

Let us consider a simple—perhaps the simplest—example of such

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a situation: the Bénard problem. When a fluid layer is heated from below, the system undergoes a top-heavy arrangement which is potentially unstable (provided that the fluid has a positive thermal expansion coefficient). Therefore the fluid has a tendency to redistribute itself. As a consequence the temperature gradient must exceed a certain value before the instability can manifest itself. When the temperature gradient reaches this critical value (depending on the properties of the fluid and on the thickness of the layer) convection arises which permits the fluid to adjust. Convection however begins in a peculiar way, in that a stationary instability occurs, which results in an arrangement of convection cells or convection rolls, first demonstrated by Bénard in 1900.(3) The phenomenon described above has been studied by a number of workers, and particularly by Lord Rayleigh, Bénard, and Chandrasekhar, who gives a detailed treatment of the linearized problem.(4) Thanks to these and other nonlinear studies,(5) the Bénard instability is quite well described and understood from the point of view of the macroscopic theory. The question now arises as to what are the dynamics of the fluctuations leading to the convective instability. In order to explore this difficult question, one would like to probe the density fluctuations in the fluid which experiences an increasing temperature gradient until it reaches its critical value. Such a procedure could elucidate the evolution of different normal modes of the fluid and determine if they are increasingly affected while approaching the " Bénard critical point ". If so the onset of the convective instability would bear striking resemblance to second order phase transitions which are triggered by " soft modes ".

In addition to the previous unanswered theoretical question regarding the evolution of the density fluctuations from the equilibrium state up to the vicinity of the " instability critical point ", one is also interested in how the problem might be studied from an experimental viewpoint. For example, an experimental study could determine whether—and if so, when—classical hydrodynamics becomes invalid in treating density fluctuations in a system submitted to an external force.

Usually light scattering appears as a very convenient tool for obtaining the power-spectrum of the density–density correlations. In the present case, because the modes which will be mostly affected by the process are long wave modes (indeed the " soft mode " wavelength is of order of the fluid layer thickness, i.e., $\lambda \sim 1$ mm) experiments should be performed in the very near forward direction ($k = 2k_0 \sin \theta/2$, where θ is the scattering angle and k_0, the wavenumber of the incident light). This certainly represents a non-trivial technical difficulty, as probing the " soft mode " requires a scattering angle $\sim 10^{-4}$ radian. However as other modes of the fluid are expected to be also affected, it appears more easily feasible to look at those modes with the shortest wavenumber experimentally accessible. The question now is how are the modes of the fluid modified by the presence of an external field. To attempt to answer this question, we present in Sec. 2 a hydrodynamic theory for a binary system to obtain the linearized equations governing the evolution of the fluctuations when such a system is subject to an external linear temperature gradient.

From the set of linearized equations, we derive, in Sec. 3, the dispersion equation, which is solved to determine the characteristic modes of the system. The last section is devoted to a discussion of the modifications these modes undergo when the system passes from the stability regime to the instability regime. The possibility of investigating such effects experimentally is also discussed.

2. Hydrodynamic Theory

Consider a binary system subject to an external force **F**. The hydrodynamic equations read:(4,6)

Continuity Equation:

$$\partial_t \rho + \nabla \cdot \rho \mathbf{v} = 0; \tag{2.1}$$

Momentum equation:

$$(\partial_t - \nu \nabla^2) \rho \mathbf{v} + \nabla p - \mathbf{F} = 0; \tag{2.2}$$

Concentration equation:

$$\partial_t \rho c + \mathbf{v} \cdot \nabla \rho c + \nabla \cdot \mathbf{i} = 0; \tag{2.3}$$

Energy equation:

$$\partial_t s + \mathbf{v} \cdot \nabla s + \frac{1}{\rho T} \nabla \cdot (\mathbf{q} - \mu \mathbf{i}) + \frac{1}{\rho T} \mathbf{i} \cdot \nabla \mu = 0. \tag{2.4}$$

Here ρ is the density; $\mathbf{v}$, the velocity; ν, the kinematic shear viscosity; p, the pressure; c, the concentration; s, the entropy density; T, the temperature; and μ, the chemical potential. The external force, $\mathbf{F}$, here is

$$\mathbf{F} = -g\rho\boldsymbol{\lambda}; \ \boldsymbol{\lambda} = (0,0,1), \tag{2.5}$$

where g is the gravity constant.

The diffusion flux, $\mathbf{i}$, and the heat flux, $\mathbf{q}$, are given respectively by

$$\mathbf{i} = -\rho D\left[\nabla c + c(1-c)\frac{D'}{D}\nabla T + c(1-c)\frac{D''}{D}\nabla p\right], \tag{2.6}$$

$$\mathbf{q} = T\left[c(1-c)\frac{D'}{D}\mu_c - \mu_T\right]\mathbf{i} + \mu\mathbf{i} - \kappa\nabla T, \tag{2.7}$$

with

$$c(1-c)\frac{D'}{D} = \frac{k_T}{T} = \frac{\mu_T}{\mu_c} + \frac{\xi}{\mu_c}, \tag{2.8}$$

$$c(1-c)\frac{D''}{D} = \frac{k_p}{p} = \frac{\mu_p}{\mu_c} = -\frac{\gamma_c/\rho}{\mu_c}, \tag{2.9}$$

where

$$\mu_x = \left.\frac{\partial\mu}{\partial x}\right|_{y,z}; \ x,y,z = p,T,c, \tag{2.10}$$

and

$$\gamma_c = \left.\frac{1}{\rho}\frac{\partial\rho}{\partial c}\right|_{p,T}. \tag{2.11}$$

Here κ is the thermal conductivity coefficient; D, the diffusion coefficient; D', the thermal diffusion coefficient; and D'', the barodiffusion coefficient. The dimensionless quantity k_T is called the thermal diffusion ratio and is not independent of the transport properties of the fluid, since ξ which appears in Eq. (2.8) is defined through the diffusion flux,

$$\mathbf{i} = -\frac{\rho D}{\mu_c}(\nabla\mu + \xi\nabla T). \tag{2.12}$$

On the other hand, it is seen from Eq. (2.9) that the dimensionless quantity k_p is entirely determined by thermodynamic properties above, and is usually a small quantity. Therefore unless there is a considerable pressure gradient in the fluid, the last term in Eq. (2.6) may be neglected. For the sake of completeness, we shall keep all terms in the present treatment and omit those which appear as small quantities only when approximations are in order. We now define the physical conditions of the system: a fluid layer of thickness d (chosen in the z direction, whereas the dimensions of the system are infinite in the x,y plane) is heated from below in such a way that the system is subject to a linear adverse temperature gradient. Furthermore, we consider the case of a dilute system ($c \ll 1$). Then from the hydrodynamic equations, the steady state is defined by

$$\nabla T^s = -\beta\boldsymbol{\lambda}; \tag{2.13}$$

$$\nabla p^s = -g\rho_0\boldsymbol{\lambda}, \tag{2.14}$$

$$\nabla c^s = \boldsymbol{\lambda}\Delta c^s \cong c_0\left(\beta\frac{D'}{D}+g\rho_0\frac{D''}{D}\right)\boldsymbol{\lambda}, \tag{2.15}$$

where the subscript 0 refers to the initial equilibrium state (reference state).

First order perturbation around the steady state yields

$$\rho = \rho^s + \left.\frac{\partial\rho}{\partial p}\right|^s \delta p + \left.\frac{\partial\rho}{\partial c}\right|^s \delta c + \left.\frac{\partial\rho}{\partial T}\right|^s \delta T \tag{2.16}$$

where

$$\rho^s = \rho_0\left[1-\alpha(T^s-T_0)-\frac{\gamma}{u_0^2}(p^s-p_0)+\gamma_c(c^s-c_0)\right], \tag{2.17}$$

with α, the thermal expansion coefficient,.

$$\alpha = -\frac{1}{\rho_0}\left.\frac{\partial\rho}{\partial T}\right|_0; \tag{2.18}$$

γ, the specific heat ratio,

$$\gamma = C_p/C_v; \tag{2.19}$$

and u_0, the sound velocity ($\sim 10^5$ cm sec^{-1}). Neglecting small terms ($\ll \alpha \sim 10^{-3}$, i.e. the terms in the expansion which are proportional to α^2 and those proportional to u_0^{-2}) we obtain the density fluctuation as given by$^{(6)}$

$$\delta\rho = -\alpha\rho_0\delta T + \gamma\rho_0\delta c. \tag{2.20}$$

We now proceed to the linearization of the hydrodynamic equations.

The procedure is straightforward for Eqs. (2.1) and (2.2), as we obtain for the linearized continuity equation

$$\partial_t \delta\rho + \rho_0 \nabla \cdot \mathbf{v} = 0, \tag{2.21}$$

and for the linearized momentum equation

$$(\partial_t - \nu\nabla^2)\rho_0 \mathbf{v} + \nabla\delta p + g\boldsymbol{\lambda}\delta\rho = 0. \tag{2.22}$$

Linearizing the concentration equation yields

$$\partial_t \delta c + \mathbf{v} \cdot \nabla c^s - DA = 0, \tag{2.23}$$

$$\begin{aligned} A = {} & \nabla^2\delta c + c^s \frac{D'}{D} \nabla^2 \delta T + c^s \frac{D''}{D} \nabla^2 \delta p \\ & + \frac{D'}{D} (\nabla c^s \cdot \nabla \delta T + \nabla T^s \cdot \nabla \delta c) + \frac{D''}{D} (\nabla c^2 \cdot \nabla \delta p + \nabla p^s \cdot \nabla \delta c). \end{aligned} \tag{2.24}$$

Noticing that

$$c^s = c_0(1 + \mathcal{O}(k_T, k_p)) ; \ \ \nabla c^s = \mathcal{O}(k_T, k_p), \tag{2.25}$$

where k_T and k_p are small quantities ($\lesssim 10^{-3}$), we retain only first order terms in k_T and k_p to obtain.

$$\begin{aligned} & \left[\partial_t - D\left(\nabla^2 - \frac{\nabla c^s}{c_0} \cdot \nabla\right)\right] \delta c + \mathbf{v} \cdot \nabla c^s \\ & \qquad - c_0(D'\nabla^2 \delta T + D''\nabla^2 \delta p) = 0. \end{aligned} \tag{2.26}$$

Before linearizing the energy equation, we want to make a change of thermodynamic variables by using the identity[8]

$$\begin{aligned} \partial_t s + \mathbf{v} \cdot \nabla s = {} & \frac{C_v}{T}\left[(\gamma - 1)\frac{\nabla \cdot \mathbf{v}}{\alpha} + \partial_t T + \mathbf{v} \cdot \nabla T\right] \\ & - \mu_T[\partial_t c + \mathbf{v} \cdot \nabla c]. \end{aligned} \tag{2.27}$$

Combination of Eqs. (2.2), (2.4), (2.6), (2.7), and (2.27) yields after a few manipulations

$$\begin{aligned} & C_v\left(\partial_t T + \mathbf{v} \cdot \nabla T + (\gamma - 1)\frac{\nabla \cdot \mathbf{v}}{\alpha}\right) - \mu_c T_c \frac{D'}{D}(\partial_t c + \mathbf{v} \cdot \nabla c) \\ & = \frac{\kappa}{\rho_0}\nabla^2 T - \frac{1}{\rho_0}\left(c\frac{D'}{D}\mu_c - \mu_T\right)\mathbf{i} \cdot \nabla T - \frac{T}{\rho_0}\frac{D'}{D}\mu_c \mathbf{i} \cdot \nabla c - \frac{1}{\rho_0}\mathbf{i} \cdot \nabla\mu, \end{aligned} \tag{2.28}$$

with

$$\nabla\mu = \mu_c \nabla c + \mu_T \nabla T + c\frac{D''}{D}\mu_c \nabla p. \tag{2.29}$$

Eq. (2.28) is now linearized. Accounting for the fact that only first order terms in k_T, and k_p are retained, we obtain

$$C_v\left[\partial_t\delta T+\mathbf{v}\cdot\nabla T^s+(\gamma-1)\frac{\nabla\cdot\mathbf{v}}{\alpha}\right]$$

$$=\frac{\kappa}{p_0}\nabla^2\delta T+\mu_c k_T D\left[\nabla^2\delta c+\frac{k_T}{T}\nabla^2\delta T+\frac{k_p}{p}\nabla^2\delta p\right]. \qquad (2.30)$$

Now using the continuity equation (2.21) and the equation of state (2.20), one has

$$\frac{1}{\alpha}\nabla\cdot\mathbf{v}=-\frac{1}{\alpha\rho_0}\partial_t\delta\rho=\partial_t\delta T-\frac{\gamma_c}{\alpha}\partial_t\delta c, \qquad (2.31)$$

which is substituted into Eq. (2.30) to yield

$$\partial_t\delta T+\frac{1}{\gamma}\mathbf{v}\cdot\nabla T^s-\frac{\gamma-1}{\gamma}\frac{\gamma_c}{\alpha}\partial_t\delta c$$

$$=\lambda'\nabla^2\delta T-\frac{\mu_c k_T}{C_p}\left(\partial_t\delta c+\mathbf{v}\cdot\nabla c^s+\frac{D}{c_0}\nabla c^s\cdot\nabla\delta c\right) \qquad (2.32)$$

where

$$\lambda'=\kappa/\rho_0 C_p \qquad (2.33)$$

is the thermal diffusivity coefficient.

Now, substituting the equation of state (2.20) into the Navier–Stokes equation (2.22), and taking twice the curl ($\nabla\times\nabla\times$) of the result, we obtain

$$(\partial_t-\nabla\nabla^2)\nabla^2 w-g(\nabla^2-\partial_z{}^2)(\alpha\delta T-\gamma_c\delta c)=0, \qquad (2.34)$$

where w is the z-component of the velocity $\mathbf{v}$. To conclude this section, let us summarize the set of linearized hydrodynamic equations we have obtained for a two-component fluid layer subject to an external linear temperature gradient. With the definitions

$$d_z=\rho_0 w;\ d_T=\rho_0\delta T;\ d_c=\rho_0\delta c, \qquad (2.35)$$

we have

$$(\partial_t-\nu\nabla^2)\nabla^2 d_z-g(\nabla^2-\partial_z{}^2)(\alpha d_z-\gamma_c d_c)=0, \qquad (2.36a)$$

$$\left[\partial_t - D\left(\nabla^2 - \frac{\Delta c^s}{c_0}\,\partial_z\right)\right]d_c + \Delta c^s d_z$$

$$- D\nabla^2\left(\frac{k_T}{T_0}\,d_T + \frac{k_p}{\rho_0}\,\rho_0\delta p\right) = 0, \qquad (2.36\text{b})$$

$$\partial_t d_T - \frac{\beta}{\gamma}\,d_z - \frac{\gamma - 1}{\gamma}\,\frac{\gamma_c}{\alpha}\,\partial_t d_c - \lambda'\nabla^2 d_T$$

$$- \frac{\mu_c k_T}{C_p}\left(\partial_t d_c + \Delta c^s d_z + D\,\frac{\Delta c^s}{c_0}\,\partial_z d_c\right) = 0 \qquad (2.36\text{c})$$

Notice that in the absence of external force (i.e. $\beta = 0$, $\Delta c^s = 0$) one retrieves the usual hydrodynamic equations for a binary system at rest.⁽⁹⁾

3. Instability " Soft Modes "

The set of linearized equations (2.36) is most easily solved by Laplace–Fourier transformation. In performing the transformation, we introduce two approximations: k_p being a small quantity we shall take the limit $k_p \to 0$, and set $\gamma_c = 0$ in Eqs. (2.36); second, we shall neglect terms proportional to $\mathbf{k}$ (or k_z) as being several orders of magnitude smaller than the other terms ($\propto k^2$). We then obtain the set

$$(s + \nu k^2)k^2\tilde{d}_z(k,s) - \alpha g(k^2 - k_z^{\,2})\tilde{d}_T(k,s) = k^2 d_z(k,0), \qquad (3.1\text{a})$$

$$\Delta c^s\tilde{d}_z(k,s) + D\,\frac{k_T}{T_0}\,k^2\tilde{d}_T(k,s)$$

$$+ (s + Dk^2)\tilde{d}_c(k,s) = d_c(k,0),$$

$$-\left(\frac{\beta}{\gamma} + \frac{\mu_c k_T}{C_p}\Delta c^s\right)\tilde{d}_z(k,s) + (s + \lambda' k^2)\tilde{d}_T(k,s)$$

$$+ \frac{\mu_c k_T}{T}\,s\tilde{d}_c(k,s) = d_T(k,0) - \frac{\mu_c k_T}{C_p}\,d_c(k,0), \qquad (3.1\text{c})$$

where s is the complex Laplace variable ($s = \epsilon + i\omega$) and the tilda denotes the Laplace transform. Notice that because of the first approximation, ($\gamma_c = 0$), Δc^s, Eq. (2.15), reduces here to

$$\Delta c^s \simeq \beta\frac{k_T}{T_0}. \qquad (3.2)$$

From the above set of equations, (3.1), we derive the dispersion equation, which reads

$$\begin{bmatrix} (s+\nu k^2)k^2 & -\alpha g(k^2-k_z{}^2) & 0 \\ \Delta c^s & Dk^2\dfrac{k_T}{T_0} & (s+Dk^2) \\ -\left(\dfrac{\beta}{\gamma}+\dfrac{\mu_c k_T}{C_p}\Delta C^s\right) & (s+\lambda' k^2) & -s\dfrac{\mu_c}{C_p}k_T \end{bmatrix} = 0 \qquad (3.3)$$

or

$$(s+\nu k^2)(s+\lambda' k^2)(s+Dk^2)+s(s+\nu k^2)\Delta k^2-(s+\mathscr{D}k^2)\nu k^2\lambda' k^2\mathscr{R}=0, \quad (3.4)$$

where

$$\mathscr{D}=D\left(1+\frac{\mu_c k_T{}^2}{C_p T_0}\right)=D+\Delta, \qquad (3.5)$$

and

$$\mathscr{R}=R_a/R_c. \qquad (3.6)$$

Here R_a is the Rayleigh number defined by

$$R_a=\frac{\alpha\beta g}{\lambda\nu}d^4, \qquad (3.7)$$

with λ, the thermometric conductivity coefficient

$$\lambda=\frac{\kappa}{\rho_0 C_v}=\gamma\lambda', \qquad (3.8)$$

and R_c is the critical Rayleigh number

$$R_c=\frac{k^6 d^4}{k^2-k_z{}^2}. \qquad (3.9)$$

The boundary conditions determine the possible values of R_c. For two free surfaces, one has $k_z=n\pi/d$, so that the minimum value of R_c is

$$R_c^{\mathrm{MIN}}=\frac{(k_{xy}^2 d^2+\pi^2)^3}{k_{xy}^2 d^2}, \qquad (3.10)$$

wherefrom one finds by a standard extremum calculation that R_c is minimum for $k_{xy}^2=\pi^2/2d^2$, which corresponds to a wavelength $\lambda=2^{3/2}d$. The minimum values of R_c are⁽⁴⁾ 658 for two free surfaces, 1101 for one free surface and one rigid boundary, and 1708 for two rigid boundaries. When increasing β up to a critical value such that $R_a = R_c^{\mathrm{MIN}}$, a marginal state comes to prevail, and when R_a exceeds R_c^{MIN}, instability sets in. Hence the instability critical point corresponds to $\mathscr{R} = 1$ in the dispersion equation and our purpose is to investigate the behavior of the characteristic modes when $\mathscr{R}$ varies from 0 to 1.

Although it is always possible to solve exactly the cubic equation (3.4), it is appropriate and convenient at this point to introduce a further approximation. Since we shall consider those modes which correspond to the critical components of the power spectrum, we restrict ourselves to a frequency range such that $\omega \ll \nu k^2$, which is indeed appropriate for most usual liquids whose thermal diffusivity coefficient is one order of magnitude smaller than their coefficient of kinematic viscosity. Then Eq. (3.4) reduces to a quadratic equation whose roots read

$$s_{\pm} = -\frac{k^2}{2}[\lambda'(1-\mathscr{R}) + \mathscr{D}] \pm \frac{k^2}{2}\left\{[\lambda'(1-\mathscr{R}) + \mathscr{D}]^2 - 4\lambda' D\left[1 - \left(1 + \frac{\Delta}{D}\right)\mathscr{R}\right]\right\}^{1/2} \tag{3.11}$$

Note that when there is no external force ($\mathscr{R} = 0$) one retrieves the usual modes characteristic of a binary system at equilibrium.(9) The above result can be rewritten as

$$s_{\pm} = -\frac{k^2}{2}(\lambda'' + \mathscr{D}) \pm \frac{k^2}{2}(\lambda'' - \mathscr{D})\left[1 + \frac{4\lambda'\Delta}{(\lambda'' - \mathscr{D})^2}\right]^{1/2}, \tag{3.12}$$

with

$$\lambda'' = \lambda'(1-\mathscr{R}) \tag{3.13}$$

Now since Δ is small compared to D, and D itself is small with respect to λ', expansion of the square root in Eq. (3.12) yields in first approximation

$$s_{+} \cong -\mathscr{D}k^2 + \frac{\Delta k^2}{1-\mathscr{R}}, \tag{3.14}$$

$$s_{-} \cong -\lambda' k^2(1-\mathscr{R}) - \frac{\Delta k^2}{1-\mathscr{R}}. \tag{3.15}$$

When $k_T = 0$, i.e. in the limit of a pure fluid, one obtains from Eq. (3.12) two exactly decoupled modes: (1) the diffusion mode which is unaffected by the external force

$$s_+ = -Dk^2, \tag{3.16}$$

(where D is now the self-diffusion coefficient) and (2) the thermal diffusivity mode

$$s_- = -\lambda' k^2(1 - \mathscr{R}), \tag{3.17}$$

which in the limit $\mathscr{R} = 0$, gives the usual central mode of a pure fluid at equilibrium. These results are summarized in Table 1 and will be commented on in the next section.

Table 1

Thermal diffusivity mode	$k_T = 0$	$k_T \neq 0$
$\beta = 0$	$-\lambda' k^2$	$-\lambda' k^2 - \Delta k^2$
$\beta \neq 0$	$-\lambda' k^2(1 - \mathscr{R})$	$-\lambda' k^2(1 - \mathscr{R}) - \dfrac{\Delta k^2}{1 - \mathscr{R}}$
Diffusion mode	$k_T = 0$	$k_T \neq 0$
$\beta = 0$	$-Dk^2$	$-Dk^2\left(1 - \dfrac{\Delta}{\lambda'}\right)$
$\beta \neq 0$	$-Dk^2$	$-k^2 + \dfrac{\Delta k^2}{1 - \mathscr{R}}$

4. Discussion

Before discussing the results on the instability " soft modes ", one comment seems in order concerning the case of a binary system at equilibrium, i.e. the results given in Table 1, at $k_T \neq 0$, and $\beta = 0$. One observes that as a result of the presence of a second component in the system, the thermal diffusivity peak should be broadened by an amount $\mu_c k_T^2 D k^2 / C_p T_0$, whereas a narrowing of $\mu_c k_T^2 D^2 k^2 / \lambda' C_p T_0$ should occur for the diffusion peak. These opposite effects, although implicitly included in previous work,(9) have not been to our knowledge explicitly stated before. Although such effects are expected to be very small, it would be interesting if they could be observed experimentally.(10)

In the presence of an external temperature gradient it is the thermal diffusivity peak which will be primarily affected. Indeed, although the second terms in the RHS of Eqs. (3.14) and (3.15) should blow up when $R_a = R_c$, the effect will actually be important only when looking at the characteristic " soft mode " of wavelength $\sim d$, and will be quite negligible for the other modes since Δ is a very small quantity. Therefore we shall essentially discuss the softening of the thermal diffusivity mode, and consider the case of a pure fluid since the effect on $\lambda' k^2$ is the same as in the case of a binary system. The power spectrum of the light scattered from entropy fluctuations (or density fluctuations at constant pressure) is easily calculated from the hydrodynamic equations, once the poles are determined from the dispersion equation.$^{(11)}$ Under the conditions established in Sec. 3, i.e. for $(\lambda'/\nu)^2 \ll 1$ and $\omega \ll \nu k^2$, we obtain

$$I(k,\omega) = \left\langle \frac{\delta\rho(-k,0)\delta\rho(k,\omega)}{\delta\rho(-k,0)\delta\rho(k,0)} \right\rangle$$

$$= \frac{2\lambda' k^2(1 - R_a/R_c)}{\omega^2[1 + 2(R_a/R_c)] + (\lambda' k^2)^2(1 - R_a/R_c)^2} \tag{4.1}$$

The peak height

$$I(k,\omega = 0) = \frac{2(\lambda' k^2)^{-2}}{1 - R_a/R_c} \tag{4.2}$$

will increase with increasing β, the temperature gradient. Starting from $2(\lambda' k^2)^{-2}$, the usual value when the fluid is at equilibrium ($R_a = 0$), the peak should diverge when the marginal state is reached, i.e. when R_a is exactly equal to R_c (see Fig. 1). The half width

$$\Delta\omega = \lambda' k^2 \frac{1 - R_a/R_c}{(1 + 2R_a/R_c)^{1/2}} \tag{4.3}$$

obeys the equation

$$XY = C, \tag{4.4}$$

with

$$C = (1 + 2R_a/R_c)^{1/2}; \quad 1 \leqslant C \leqslant 3^{1/2}, \tag{4.5}$$

$$X = 1 - R_a/R_c; \quad 1 \geqslant X \geqslant 0, \tag{4.6}$$

$$Y = \lambda' k^2/\Delta\omega; \quad 1 \leqslant Y \leqslant \infty. \tag{4.7}$$

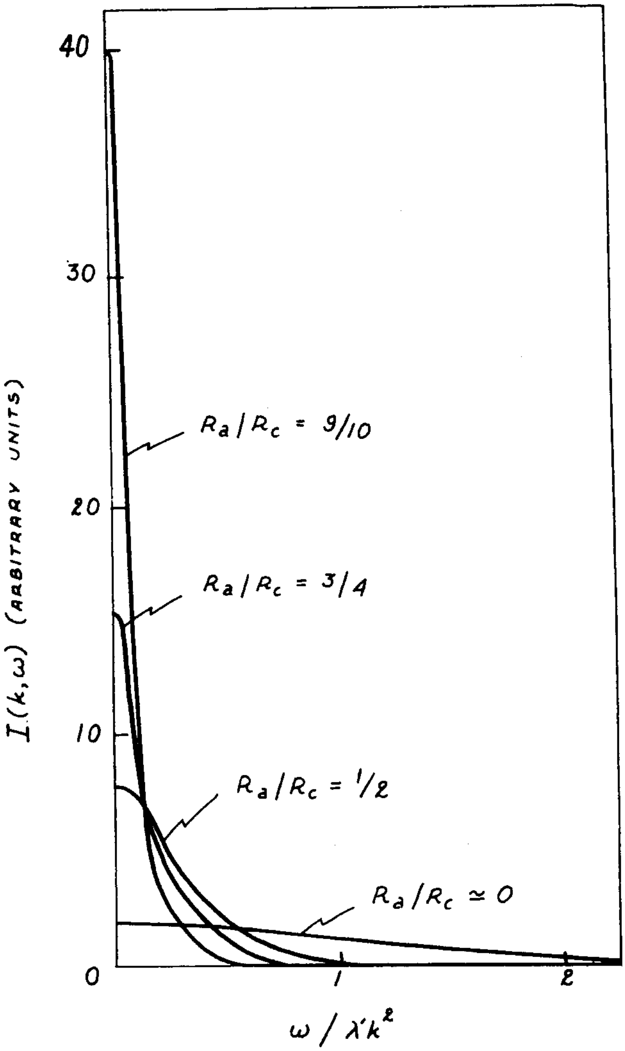

Figure 1. The power spectrum for different values of the Rayleigh number.

In other words, when increasing β, the reciprocal width grows as a hyperbola, with an asymptote at $X = 0$, as shown in Fig. 2.

An important shortcoming of the above theoretical discussion is that the spectral shape predicted in Eq. (4.1) must be scaled by the static correlation function $\langle \delta\rho(-k,0)\delta\rho(k,0)\rangle$ in order to produce the absolute scattering efficiency. This function cannot be obtained by the analysis presented here. The corresponding function at equilibrium, however, has a value sufficient to make the light scattering experiment feasible from an intensity point of view.

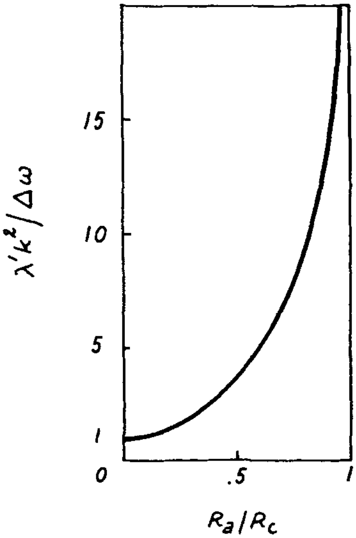

Figure 2. The reduced reciprocal half-width as a function of the Rayleigh number.

Some of the difficulties involved in the experimental observation of the onset of the instability have already been mentioned in Sec. 1. As stated before, the shorter the wavelength, the less the mode will be affected and the less important the effect will be as compared to the softening of the soft mode itself; but the easier the experiment. An additional experimental difficulty should be accounted for. Since the width is smaller for longer wavelength modes, and thus less accurately measurable, the observation of the effect—i.e. the narrowing being smaller the shorter the wavelength—may appear as a delicate operation. The feasibility of the experiment will therefore depend on obtaining a good compromise between these difficulties.

One final remark seems in order. The smallness of the effect in a pure fluid (or in a binary mixture) is connected with the value of the thermal diffusivity. A possible way around this obstacle is to consider a Brownian system. Indeed utilizing particles in suspension to probe the density fluctuations would have the effect of replacing $\kappa/\rho_0 C_p$ by the diffusion constant, D, of the Brownian particles, which is smaller by several orders of magnitude than the thermal diffusivity. If for a Brownian system, there exists a corresponding Rayleigh number where $\lambda\nu$ appearing in the denominator should be replaced by D^2, then the effect would be significantly larger even for larger wavenumber modes (e.g. $10^4 < k < 10^5$ for a suspension of polystyrene balls in water, with a diameter $\sim 10^{-5}$ cm and $D \sim 10^{-8}$ cm^2 sec^{-1}). Furthermore the scattering from the Brownian particles is much more intense than the scattering from thermal fluctuations.

We believe that such effects as predicted in the present work should be observable with the presently available techniques. Such procedures would then comprise an interesting tool to investigate thermal instabilities in fluids, as it has proved to be in the case of hydrodynamic instabilities.(12)

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Addendum

At the time the present work was completed, the author was not aware of the work by V. M. Zaitsev and M. I. Shliomis (*Soviet Physics J.E.T.P.* **32,** 866 (1971)) who, by a different method, are led essentially to the same conclusions.

However, essential differences between the Russian work and ours should be mentioned. Zaitsev and Shliomis treat the problem on the basis of the hydrodynamic fluctuations theory by introducing the concept of fluctuation velocities and temperatures excited by random forces. Therefore their starting set of equations differs from ours by the presence of " injected " random terms, i.e. an " extraneous stress tensor " in the momentum equation, and an " extraneous heat flux " in the energy equation. Following Landau and Lifshitz (*Soviet Physics J.E.T.P.* **5**, 511, 1957) an anzatz is introduced for the autocorrelation of the extraneous forces and the equations are then solved by the eigenfunctions expansion method.

This allows them to treat the problem for a single-component system with arbitrary geometry and rigid boundaries at the cost of mathematical complexity, whereas our method presents the advantage of analytical simplicity with the restriction to systems with free boundaries. Zaitsev and Shliomis' main goal is the analytical investigation of the behavior of the hydrodynamic fluctuations near convection threshold. In the present paper we essentially develop a simple method indicating the possibility of a light scattering experiment to probe the onset of the convective instability.

Most important is the conclusion that the Russian work and the present paper confirm the belief that a divergence of the characteristic fluctuations power spectrum should occur at the convection threshold. Therefore in the immediate vicinity of the instability critical point, the linear treatment used in both papers should be expected to become invalid. It should also be mentioned that the brackets in Eq. (4.1) of the present paper, as well as in Zaitsev and Shliomis' work (see footnote 2 in their paper), denote an ensemble average over the steady state of the system. Although such a procedure is strictly valid only for a system at thermodynamic equilibrium, the extension of the averaging procedure should be applicable to the present case, since local thermodynamic equilibrium is assumed. However, when deviations from equilibrium are no longer small, the procedure becomes questionable, as in the *immediate* vicinity of the convection threshold.

It is important to notice that the analogy between the phenomenon considered herein and a second order phase transition (as suggested in the present paper and in the Russian article) is essentially *phenomenological* to the extent that the " soft mode " picture provides a satisfactory description in both cases. The analogy has to be understood in terms of a translation from the molecular description language used for phase transitions to the hydrodynamic mode language used for instability phenomena.

On the other hand it is worthwhile to mention that at the level of a *microscopic* analysis there is presently no evidence for an analogy between hydrodynamic or thermal instabilities and phase transitions for the very reason that the microscopic mechanism governing the evolution of a system towards an instability point remains at present a totally open question.

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