

## Hydrodynamic modes and light scattering near the convective instability

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An analysis is presented of the behavior of the hydrodynamic modes in a horizontal fluid layer subject to a downward-directed temperature gradient which, when reaching a critical value, drives the system into convective instability. It is found that the combination of this thermal constraint and gravity gives rise to a coupling between the heat-diffusion mode and the transverse mode  $(\text{curl curl } \vec{v})_z$ , with  $\vec{v}$ , the velocity field, and the  $z$  axis pointing in the vertical direction. As a result of this mode coupling, which is absent in a fluid at equilibrium, the damping factor of one of the coupled modes goes to zero when the temperature gradient increases to its critical value. On the basis of this mode analysis, the spectral distribution of the light scattered by the nonequilibrium fluid is then computed. The main features of the light-scattering spectrum consist of the appearance of an additional central component and the respective narrowing and broadening of the spectral components corresponding to the coupled modes as the instability critical point is approached.

### I. INTRODUCTION

Consider a horizontal fluid layer in which a linear downward-directed temperature gradient is maintained. Stationary convection sets on spontaneously in the fluid layer when the temperature gradient reaches a critical value. This example constitutes one of the simplest cases of a hydrodynamic instability and is often referred to as the Bénard problem in classical physics.<sup>1</sup> The question of the stability criteria in such systems, as well as the determination of the critical value of the parameters characterizing the transition between the stability region and the instability domain, has been studied for a long time.<sup>1</sup> However, it is only recently that the problem of the dynamics of fluctuations in such systems has received attention.<sup>2-4</sup> In the present paper, we investigate, on the basis of linear hydrodynamic theory, how the hydrodynamic modes characterizing the dynamics of fluctuations in a fluid are affected by the presence of the "external force" resulting from the combination of an imposed temperature gradient and gravity.

Section II is devoted to a description of the system considered, for which the corresponding set of linearized hydrodynamic equations for the fluctuations in the steady state is given. In Sec. III we present a treatment of the problem based on the analysis of the structure of the hydrodynamic matrix, an approach which possesses the advantage of analytical simplicity. The analysis is performed by comparison with the structure of the hydrodynamic matrix for the fluid in the equilib-

rium state in order to exhibit the modifications of the normal modes due to the external force. In Sec. IV we apply this mode analysis to the computation of the spectral distribution of the light scattered from the nonequilibrium fluid. Indeed, since the dynamics of fluctuations can be probed experimentally by light-scattering spectroscopy, information on the dynamics of the fluid evolving towards the instability critical point can be obtained from the light-scattering spectrum. A discussion of the spectral features is given in Sec. V. The present analysis predicts the appearance of a new central mode when the fluid departs from its equilibrium state. Furthermore, one finds that one of the two central modes behaves as a "soft mode"<sup>5</sup>; this follows from the observation<sup>2,4</sup> that the correlation time of the corresponding thermal fluctuations diverges near convection threshold as  $(R_c - R)^{-1}$ , where  $R$  is the Rayleigh number (the dimensionless temperature gradient) and  $R_c$  its critical value. This result seems to indicate an analogy between the behavior of a fluid at the instability threshold and the situation encountered in structural phase transitions.<sup>5</sup>

### II. LINEARIZED HYDRODYNAMIC EQUATIONS

The steady state of the fluid in the stability region is characterized as follows: a linear downward-directed temperature gradient is maintained steadily, the macroscopic fluid velocity is zero, and the gravitational force is balanced by the hydrostatic pressure gradient. Labeling the steady-state variables with superscript  $s$ , one

has

$$T^s = T_0 - \beta z, \quad \vec{v}^s = 0, \quad \text{grad} \phi^s = -g \rho^s \hat{n}. \quad (1)$$

Here the subscript 0 denotes the value of the corresponding quantity at the reference position (taken here at the lower boundary  $z=0$ ),  $\beta$  is the value of the temperature gradient,  $g$  is the gravitational constant, and  $\hat{n}$  is a unit vector pointing in the positive  $z$  direction.

The linearized hydrodynamic equations for the fluctuations in the steady state read<sup>6</sup>

$$\begin{aligned} \frac{\partial \delta \rho}{\partial t} &= -\rho_0 \text{div} \vec{v} + \beta v_z \left( \frac{\partial \rho}{\partial T} \right)_p, \\ \frac{\partial \vec{v}}{\partial t} &= -\frac{1}{\rho_0} \text{grad} \delta p + \frac{\eta}{\rho_0} \nabla^2 \vec{v} \\ &\quad + \frac{\zeta + \frac{1}{3}\eta}{\rho_0} \text{grad} \text{div} \vec{v} - g \hat{n} \frac{\delta \rho}{\rho_0}, \end{aligned} \quad (2)$$

$$\frac{\partial \delta s}{\partial t} - \frac{\beta c_p v_z}{T_0} = \frac{\kappa}{\rho_0 T_0} \nabla^2 \delta T.$$

Here  $\delta \rho$ ,  $\delta T$ ,  $\delta s$ , and  $\vec{v}$  are fluctuations in the corresponding steady-state variables, e.g.,  $\rho = \rho^s + \delta \rho$ . To arrive at Eqs. (2), the following approximations have been made: the transport coefficients ( $\eta$ ,  $\zeta$ , and  $\kappa$ ) and the specific heat at constant pressure ( $c_p$ ) are taken as constants; after linearization of the hydrodynamic equations, the quantities  $\rho^s$  and  $T^s$  have been approximated by  $\rho_0$  and  $T_0$ ; in the first and third equation of Eqs. (2), the terms  $\rho_0 g v_z (\partial \rho / \partial p) \ll -\beta v_z (\partial \rho / \partial T)_p$  and  $-\rho_0^{-1} g v_z \times (\partial \rho / \partial T)_p \ll \beta c_p v_z / T_0$ , respectively, have been omitted. Equations (2) constitute the basic set of equations for our investigation of the behavior of the hydrodynamic normal modes under the influence of the external force defined above and manifesting itself by the presence of terms containing either  $\beta$  or  $g$  in Eqs. (2).

### III. HYDRODYNAMIC NORMAL MODES

For the sake of further comparative analysis, let us briefly recall the hydrodynamic modes in an equilibrium fluid.<sup>7</sup> One notes first that in an isotropic medium, the velocity field can be specified by the variables  $\text{div} \vec{v}$  and two components of  $\text{curl} \vec{v}$ . Alternatively, the following choice is also possible:  $\text{div} \vec{v}$ ,  $(\text{curl} \vec{v})_z$  and  $(\text{curl} \text{curl} \vec{v})_z$ , variables which will appear to be particularly appropriate below. For a fluid at equilibrium, one finds three longitudinal modes: the heat diffusion mode  $\delta s$  and two sound modes which are linear combinations of  $\delta p$  and  $\text{div} \vec{v}$ . Furthermore, there are two transverse modes defined by two of the compo-

nents of  $\text{curl} \vec{v}$ , or by  $(\text{curl} \vec{v})_x$  and  $(\text{curl} \text{curl} \vec{v})_z$ .

When the fluid is subject to an external force oriented in a preferential direction, there is isotropy breaking of the system, which renders the above separation into longitudinal (irrotational) and transverse (rotational) components invalid. Indeed, as described in Sec. I, the system considered here exhibits the following symmetry: rotational invariance with respect to the  $z$  axis, inversion invariance in the  $(x, y)$  plane, and reflection invariance with respect to planes containing the  $z$  axis. Therefore a convenient description of the velocity field is the following<sup>1</sup>: the horizontal component of the velocity is decomposed into an irrotational part and a rotational part

$$\phi = \frac{\partial v_x}{\partial x} + \frac{\partial v_y}{\partial y}$$

and

$$\psi = \frac{\partial v_y}{\partial x} - \frac{\partial v_x}{\partial y} [= (\text{curl} \vec{v})_z],$$

respectively, while the vertical component of the velocity,  $v_z$ , is used unchanged. The variables  $\phi$  and  $v_z$  transform as scalars and can thus couple to the thermodynamic variables, while  $\psi$  cannot couple to any of the other variables, as it changes sign under reflections with respect to planes containing the  $z$  axis. Since the following functional of  $\phi$  and  $v_z$ ,

$$\xi = \frac{\partial \phi}{\partial z} - \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) v_z$$

is just  $(\text{curl} \text{curl} \vec{v})_z$ , one sees that the velocity field is also defined by the set of variables  $\text{div} \vec{v}$ ,  $\psi$ , and  $\xi$ , which, as mentioned above, is also appropriate for the description of the velocity field of the fluid at equilibrium. In this notation,  $\psi$  and  $\xi$  are the two transverse modes. It is now a matter of simple algebra to write the set of Eqs. (2) for the spatial Fourier components of the new variables  $\delta s$ ,  $\delta p$ ,  $\varphi = \text{div} \vec{v}$ ,  $\xi$ , and  $\psi$ . For a fluid at equilibrium [i.e., omitting the terms containing  $\beta$  and  $g$  in Eqs. (2)], one obtains

$$\frac{\partial}{\partial t} \begin{bmatrix} \delta p(\vec{k}, t) \\ \varphi(\vec{k}, t) \\ \delta s(\vec{k}, t) \\ \xi(\vec{k}, t) \\ \psi(\vec{k}, t) \end{bmatrix} = -\underline{\underline{M}}^{(0)} \begin{bmatrix} \delta p(\vec{k}, t) \\ \varphi(\vec{k}, t) \\ \delta s(\vec{k}, t) \\ \xi(\vec{k}, t) \\ \psi(\vec{k}, t) \end{bmatrix}, \quad (3)$$

with

$$\underline{M}^{(0)} = \left[ \begin{array}{cc|cc} (\gamma-1)\chi k^2 & \rho_0 \left( \frac{\partial p}{\partial \rho} \right)_s & \rho_0(\gamma-1)\chi k^2/\alpha & 0 & 0 \\ -k^2/\rho_0 & (\frac{4}{3}\eta + \xi)k^2/\rho_0 & 0 & 0 & 0 \\ \hline \alpha\chi k^2/\rho_0 & 0 & \chi k^2 & 0 & 0 \\ \hline 0 & 0 & 0 & \nu k^2 & 0 \\ 0 & 0 & 0 & 0 & \nu k^2 \end{array} \right] \quad (4)$$

Here  $\chi = \kappa/(\rho_0 c_p)$  is the thermal diffusivity,  $\nu = \eta\rho_0^{-1}$  is the kinematic viscosity,  $\alpha = -(1/\rho_0)(\partial\rho/\partial T)_p$  is the thermal expansivity, and  $\gamma = c_p/c_v$ . Note that for those wave numbers which can be probed by light-scattering spectroscopy ( $k \leq 2 \times 10^5 \text{ cm}^{-1}$ ), it generally holds that  $Xk^2 \ll kc_0$ , with  $c_0 = (\partial p/\partial \rho)_s^{1/2}$ , the adiabatic sound velocity ( $\sim 10^5 \text{ cm/sec}^{-1}$ ), and  $X = \chi$  or  $(\frac{4}{3}\eta + \xi)/\rho_0$  ( $\sim 10^{-2} - 10^{-3} \text{ cm}^2/\text{sec}^{-1}$ ). Therefore, to a good approximation, the effect of the coupling terms involving  $\delta p$  and  $\delta s$  is negligible, and it follows that the characteristics of the sound modes can be determined from the upper left block of the matrix of coefficients  $\underline{M}^{(0)}$ . This matrix, which we shall refer to as the hydrodynamic matrix [the superscript (0) denoting the equilibrium state], has been partitioned into different blocks by solid and dashed lines in Eq. (4) for the sake of illustration. One has the well-known results<sup>7</sup> that the frequencies of the sound waves are given by  $\omega_s = \pm kc_0$  and their damping factor by  $\Gamma_s k^2 = \frac{1}{2}[(\frac{4}{3}\eta + \xi)/\rho_0 + (\gamma-1)\chi]k^2$ . Similarly, one finds that the damping of the heat-diffusion mode is given by  $M_{33}^{(0)} = \chi k^2$ . One also observes, as expected, that the two transverse modes with damping factors  $\nu k^2$  are completely decoupled from the other modes.

We now turn to the analysis of the nonequilibrium fluid. When the system is subject to the external force, the variables  $\delta p$ ,  $\delta s$ ,  $\phi$ , and  $v_z$

are coupled as mentioned above. Consequently, the variable  $\xi$  (which in a system at equilibrium is decoupled from the other variables) couples to  $\delta p$ ,  $\delta s$ , and  $\phi$  under the nonequilibrium conditions, while  $\psi$  remains uncoupled because of its symmetry properties. As a result of this new coupling scheme, the longitudinal modes are expected to be modified.

Starting from the set of linearized hydrodynamic equations for the fluctuations in the steady state [Eqs. (2)], we proceed along the same lines as for the analysis of the equilibrium system. The geometry of the fluid is such that the system is considered to have infinite dimensions in the  $(x, y)$  plane, and for the sake of mathematical simplicity we further consider hypothetical boundary conditions at  $z=0$  and  $z=d$  such that all variables can be Fourier transformed with respect to the three Cartesian axes, as

$$Y(\vec{r}, t) = \sum_{k_x} \int_{-\infty}^{+\infty} dk_x \int_{-\infty}^{+\infty} dk_y Y(\vec{k}, t) e^{i\vec{k} \cdot \vec{r}} \quad (5)$$

Here  $Y$  stands for any of the variables  $\delta s$ ,  $\delta p$ ,  $\phi$ ,  $\xi$ , and  $\psi$ ;  $k_x$  and  $k_y$  are continuous variables, while  $k_z$  is a discrete variable with  $k_z = k_{\perp} = 2\pi m d^{-1}$ , where  $m$  is an integer. The equation to be solved now is the same as Eq. (3), with  $\underline{M}^{(0)}$  replaced by the nonequilibrium hydrodynamic matrix  $\underline{M}$  as obtained from Eqs. (2) and given by

$$\underline{M} = \left[ \begin{array}{cc|cc} (\gamma-1)\chi k^2 & \rho_0 \left( \frac{\partial p}{\partial \rho} \right)_s & \rho_0(\gamma-1)\chi k^2/\alpha & 0 & 0 \\ -k^2/\rho_0 + g \left( \frac{\partial \rho}{\partial p} \right)_s i k_{\perp}/\rho_0 & (\frac{4}{3}\eta + \xi)k^2/\rho_0 & -g\alpha T i k_{\perp}/c_p & 0 & 0 \\ \hline \alpha\chi k^2/\rho_0 & \beta c_p i k_{\perp}/(T k^2) & \chi k^2 & -\beta c_p/(T k^2) & 0 \\ \hline g \left( \frac{\partial \rho}{\partial p} \right)_s k_{\parallel}^2/\rho_0 & 0 & -g\alpha T k_{\parallel}^2/c_p & \nu k^2 & 0 \\ \hline 0 & 0 & 0 & 0 & \nu k^2 \end{array} \right] \quad (6)$$

with  $k_{\parallel}^2 = k_x^2 + k_y^2$  and  $k^2 = k_{\parallel}^2 + k_{\perp}^2$ .

To compute the nonequilibrium normal modes, one first determines the eigenvalues of the matrix  $\underline{M}$ . As expected from symmetry considerations,

it is observed that  $\psi$  remains fully decoupled from the other variables, while the transverse mode  $\xi$  now couples to the longitudinal modes.<sup>8</sup> This situation is illustrated in Eq. (6) by the solid line

separating the lower right  $1 \times 1$  block from the upper left  $4 \times 4$  block of the hydrodynamic matrix. The latter block is further partitioned as indicated by the dashed lines, and we denote the diagonal blocks which will appear to be of primary importance in the analysis by

$$\underline{M}_I = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \quad \text{and} \quad \underline{M}_{II} = \begin{bmatrix} M_{33} & M_{34} \\ M_{43} & M_{44} \end{bmatrix}. \quad (7)$$

Indeed, under the conditions  $Xk^2 \ll kc_0$  (see above), it can be shown that the Routh-Hurwitz stability conditions<sup>9</sup> for the full hydrodynamic matrix  $\underline{M}$  effectively reduce to the stability conditions for the matrix  $\underline{M}_{II}$ , the conditions of which read

$$\text{Tr} \underline{M}_{II} > 0 \quad \text{and} \quad \det \underline{M}_{II} > 0. \quad (8)$$

The first condition is obviously fulfilled [see Eq. (6)], but it requires that  $\chi\nu k^4 > \alpha\beta g k_{\parallel}^2 k^{-2}$  to satisfy the second one. Now, this inequality can be re-written

$$R/R_c(\vec{k}) = (\alpha\beta g/\nu\chi) k_{\parallel}^2/k^8 < 1, \quad (9)$$

where  $R = \alpha\beta g d^4/\nu\chi$  is the Rayleigh number, and  $R_c(\vec{k})$  is the critical value of the Rayleigh number<sup>10</sup> for the mode specified by the wave numbers  $k_{\parallel}$  and  $k_{\perp}$ . Thus, in the region where  $R$  is smaller than its critical value (which is the domain we are investigating here), it follows from the above considerations that  $(kc_0)^2 \gg \alpha\beta g$ . Under these conditions, the elements not contained in  $\underline{M}_I$  and  $\underline{M}_{II}$  have a negligible effect on the normal modes, and the characteristic polynomial of the matrix  $\underline{M}$  is then given, to a good approximation, by

$$p(\lambda) \approx (\nu k^2 - \lambda) p_I(\lambda) p_{II}(\lambda), \quad (10)$$

where  $p_I(\lambda)$  and  $p_{II}(\lambda)$  are the characteristic polynomials of the matrices  $\underline{M}_I$  and  $\underline{M}_{II}$ , respectively. In the matrix  $\underline{M}_I$ , the term containing  $g$  is much smaller than the usual "equilibrium term." Therefore the sound modes remain effectively unchanged by the presence of the external force. However, the situation is quite different when one analyzes the matrix  $\underline{M}_{II}$ —i.e., for the heat-diffusion mode and the transverse mode  $\xi$ . The damping factors of these modes are obtained from the equation  $p_{II}(\lambda) = 0$ , which yields the eigenvalues

$$\lambda_{\pm} = \frac{k^2}{2} \left[ (\nu + \chi) \pm \left( (\nu + \chi)^2 - 4\nu\chi \frac{R_c(\vec{k}) - R}{R_c(\vec{k})} \right)^{1/2} \right]. \quad (11)$$

Equation (11) expresses the effect of the coupling between the heat-diffusion mode and the transverse mode  $\xi$  due to the external force. One observes that their eigenvalues reduce to  $\lambda_{\pm}^{(0)} = \nu k^2$  and  $\lambda_{\pm}^{(0)} = \chi k^2$  when  $R = 0$  (i.e., one retrieves the

usual results for the fluid at equilibrium). Because in most fluids  $\nu$  is larger than  $\chi$ , it follows from Eq. (11) that the damping of the heat-diffusion mode will decrease, while the damping of the vorticity mode will increase, as the temperature gradient increases. In particular, when the instability critical point is approached, one finds that  $\lambda_{-} \rightarrow 0$  and  $\lambda_{+} \rightarrow (\nu + \chi)k^2$ .

The coupling also induces changes in the structure of the normal modes, i.e., in those combinations of  $\delta s(\vec{k}, t)$  and  $\xi(\vec{k}, t)$  which exhibit the average time behavior

$$\langle \gamma_{\pm}(\vec{k}, t) \rangle = \gamma_{\pm}(\vec{k}, t) e^{-\lambda_{\pm} t}.$$

Explicit computation yields

$$\begin{aligned} \gamma_{+}(\vec{k}, t) &= - \left( \frac{\alpha g T^2 k^2 k_{\parallel}^2}{\beta c_p^2} \right)^{1/2} \left( \frac{\chi k^2 - \lambda_{-}}{\chi k^2 + \nu k^2 - 2\lambda_{-}} \right)^{1/2} \\ &\quad \times \delta s(\vec{k}, t) + \left( \frac{\nu k^2 - \lambda_{-}}{\chi k^2 + \nu k^2 - 2\lambda_{-}} \right)^{1/2} \xi(\vec{k}, t), \\ \gamma_{-}(\vec{k}, t) &= \left( \frac{\nu k^2 - \lambda_{-}}{\chi k^2 + \nu k^2 - 2\lambda_{-}} \right)^{1/2} \delta s(\vec{k}, t) \\ &\quad + \left( \frac{\beta c_p^2}{\alpha g T^2 k^2 k_{\parallel}^2} \right)^{1/2} \left( \frac{\chi k^2 - \lambda_{-}}{\chi k^2 + \nu k^2 - 2\lambda_{-}} \right)^{1/2} \xi(\vec{k}, t). \end{aligned} \quad (12)$$

In the limit  $\beta \rightarrow 0$ , it follows from Eq. (11) that  $\lambda_{-} \rightarrow \chi k^2$ , and it is clear from the above expressions that  $\gamma_{+}(\vec{k}, t) \rightarrow \xi(\vec{k}, t)$  and  $\gamma_{-}(\vec{k}, t) \rightarrow \delta s(\vec{k}, t)$ —i.e., one retrieves the normal modes for the equilibrium fluid.

#### IV. LIGHT-SCATTERING SPECTRUM

Because the dynamics of fluctuations in a fluid can be probed by light-scattering spectroscopy, and since, as seen above, the hydrodynamic normal modes are modified when the fluid is subject to an imposed temperature gradient, we now compute the spectral distribution of the scattered light and discuss the ensuing changes in the light-scattering spectrum.<sup>3,11</sup> The latter is proportional to the spectral density of the  $\vec{k}$ th spatial Fourier component of the fluctuations in the optical dielectric constant  $\delta\epsilon$ , which in turn can be expressed in terms of the thermodynamic fluctuations. Thus the intensity of the scattered light can be written<sup>12</sup>

$$I(\vec{k}, \omega) \propto \left( \frac{\partial \epsilon}{\partial s} \right)_p^2 \langle |\delta s(\vec{k}, \omega)|^2 \rangle + \left( \frac{\partial \epsilon}{\partial p} \right)_s^2 \langle |\delta p(\vec{k}, \omega)|^2 \rangle, \quad (13)$$

where we have omitted the cross terms of the type  $\langle \delta s^*(\vec{k}, \omega) \delta p(\vec{k}, \omega) \rangle$ , as they are negligible under

the condition  $Xk^2 \ll kc_0$ . The second term on the right-hand side of Eq. (13) represents the spectral contribution of the sound modes, which, as seen in Sec. III, are essentially unaffected by the non-equilibrium conditions. We shall therefore restrict our analysis to the behavior of the spectral density of the entropy fluctuations, which undergoes significant changes under the influence of the external force. Because in this case  $\delta s$  relaxes into two normal modes [see Eq. (12)], it is expected that the central line of the light-scattering spectrum will consist of two Lorentzians. By a standard calculation one indeed obtains

$$I^{\text{central}}(\vec{k}, \omega) \propto \left( \frac{\partial \epsilon}{\partial s} \right)_p^2 \langle |\delta s(\vec{k})|^2 \rangle \\ \times \left[ \left( \frac{\nu k^2 - \lambda_-}{\chi k^2 + \nu k^2 - 2\lambda_-} \right) \frac{\lambda_-}{\lambda_-^2 + \omega^2} \right. \\ \left. + \left( \frac{\chi k^2 - \lambda_+}{\chi k^2 + \nu k^2 - 2\lambda_+} \right) \frac{\lambda_+}{\lambda_+^2 + \omega^2} \right], \quad (14)$$

where  $\lambda_+$  and  $\lambda_-$  are given by Eq. (11). Here again, it is easily recognized that when  $R=0$  the second term on the right-hand side of Eq. (14) vanishes; the central line of the spectrum then reduces to a single Lorentzian (with  $\lambda_-^{(0)} = \chi k^2$ ) characteristic of the heat-diffusion mode for a fluid at equilibrium.

## V. DISCUSSION

We have presented a treatment of the problem of a horizontal fluid layer subject to an external force resulting from the combination of a thermal constraint and gravity. We have investigated the dynamics of such a system, which evolves towards convective instability when the external force increases up to a critical value.

Our treatment is based on the analysis of the structure of the hydrodynamic matrix, from which the normal modes are obtained and the light-scattering spectrum is calculated. The most important feature of the spectrum consists in the structure of the central peak, which is composed of two Lorentzians. In order to examine how the spectrum is modified by the external force, let us consider two typical cases. Considering common liquids at room temperature (e.g., toluene), typically  $\nu \sim 10\chi$ , and consequently the second term on the right-hand side of Eq. (14) can be ignored, as this spectral line will be much broader and much less intense than the heat-diffusion line. In the limit where  $\chi$  may be neglected with respect to  $\nu$ , the spectrum exhibits one single central component whose width  $\Delta\omega_- = \chi k^2 [1 - R/R_c(\vec{k})]$

narrows from its equilibrium value  $\chi k^2$  to zero when  $R$  increases from zero to its critical value  $R_c(\vec{k})$ . Consider now the case of a simple fluid (e.g., argon) where  $\nu$  and  $\chi$  are of the same order of magnitude (with  $\nu$  slightly larger than  $\chi$ ). Then, both central components should be visible in the spectrum. One predicts from Eq. (14) that, when the temperature gradient increases towards its critical value, the heat-diffusion mode narrows (as in the previous case) while the spectral line corresponding to the transverse mode broadens to the value  $(\nu + \chi)k^2$  when  $R$  goes to  $R_c(\vec{k})$ .

Now, it is clear from Eq. (11) that, since  $R/R_c(\vec{k}) \propto k^{-4}$ , the modes which will be most affected by the external force are those with small wave number. In particular, the mode which is the first one to become unstable has a wavelength on the order of the vertical dimension of the system<sup>1</sup>—i.e., typically  $\lambda_c \sim 1$  mm. As a result, the most important modifications in the spectrum are to be observed at very small scattering angles. This certainly represents a nontrivial difficulty, as probing the “critical mode” then requires a scattering angle  $\sim 10^{-4}$  rad; i.e., experiments should be performed in the very-near-forward direction. Presently available techniques in light-scattering spectroscopy should allow the probing of such modes, which are expected to exhibit dramatic changes in a fluid subject to an external force. However, as the other modes ( $\lambda < \lambda_c$ ) are also affected when the instability critical point is approached, it might appear more feasible to investigate those modes with the shortest wave number experimentally accessible. In addition, the smaller the wavelength, the broader the line-width—i.e., the easier the measure of the width—but also the smaller the effect of the narrowing near the critical point. Consequently, the most appropriate experimental conditions will depend on a good compromise between these difficulties, taking into account the performance of the experimental setup.

It is worth noting here that the first measurements of the convective-velocity field near the Bénard instability threshold [for  $R > R_c(k)$ ] have been performed very recently by Bergé and Dubois precisely by using light-scattering spectroscopy.<sup>13</sup> It should also be mentioned that some years ago Goldstein and Hagen investigated the spectral changes of Doppler-scattered light in a fluid passing from the laminar flow regime to the domain of turbulence.<sup>14</sup>

Finally, let us mention that the treatment given in the present paper is being extended to the study of the hydrodynamic modes and the light-scattering spectrum in a binary mixture heated from either below or above. An analysis of such sys-

tems based on the structure of the hydrodynamic matrix would indicate that the most important effects will arise from the coupling between the heat-diffusion mode, the transverse mode  $\xi$ , and

the concentration mode. This application of the present approach is of interest in view of the new and unexpected phenomena found in these binary systems.<sup>15</sup>

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<sup>1</sup>For an extensive discussion and basic references see S. Chandrasekar, *Hydrodynamic and Hydromagnetic Stability* (Clarendon, Oxford, 1961), Chap. 2. A recent literature survey on the subject can be found in a forthcoming article by M. G. Velarde, in *Hydrodynamics, Les Houches 1973*, edited by R. Balian (Gordon and Breach, New York, 1974).

<sup>2</sup>V. M. Zaitsev and M. I. Shliomis, *Zh. Eksp. Teor. Fiz.* **59**, 1583 (1970) [*Sov. Phys.—JETP* **32**, 866 (1971)].

<sup>3</sup>J. P. Boon, *Phys. Chem. Liq.* **3**, 157 (1972); J. P. Boon and P. Deguent, *Phys. Lett. A* **39**, 315 (1972); J. P. Boon and H. N. W. Lekkerkerker, in *Anharmonic Lattices, Structural Transitions and Melting* (Noordhoff, Leyden, The Netherlands, 1973).

<sup>4</sup>K. Kawasaki, *J. Phys. A* **6**, L4 (1973).

<sup>5</sup>H. Thomas, *IEEE Trans. Magn.* **5**, 874 (1969).

<sup>6</sup>We follow the notation used by L. D. Landau and E. M. Lifshitz, in *Fluid Mechanics* (Addison-Wesley, New York, 1958).

<sup>7</sup>See, e.g., L. P. Kadanoff and P. C. Martin, *Ann. Phys. (N. Y.)* **24**, 419 (1963).

<sup>8</sup>The modes are denoted as longitudinal and transverse in the nonequilibrium system by reference to the denomination used in the equilibrium case.

<sup>9</sup>See, e.g., P. Lancaster, *Theory of Matrices* (Academic, New York, 1969), Chap. 8.

<sup>10</sup>In the remainder, we refer to the critical Rayleigh number as the smallest value of  $R_c(\vec{k})$ .

<sup>11</sup>The spectrum of light scattered from a chemically reactive system far from equilibrium has been studied theoretically by J. M. Deutch, S. Hudson, P. J. Ortoleva, and J. Ross, *J. Chem. Phys.* **57**, 4327 (1972).

<sup>12</sup>See, e.g., G. B. Benedek in *Brandeis Lectures in Theoretical Physics 1966*, edited by M. Chretien *et al.* (Gordon and Breach, New York, 1968), Vol. 2.

<sup>13</sup>P. Bergé and M. Dubois, *Phys. Rev. Lett.* **32**, 1041 (1974).

<sup>14</sup>R. J. Goldstein and W. F. Hagen, *Phys. Fluids* **10**, 1349 (1967).

<sup>15</sup>See, e.g., P. Glansdorff and I. Prigogine, *Structure, Stability, and Fluctuations* (Wiley, New York, 1971), Chap. 11, and references contained therein.