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Kinetic Theory of Fluctuations near a Convective Instability

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The kinetic theory of density and velocity fluctuations below and above the first convective instability in a Bénard cell is presented. Results are given in a form valid for all fluid densities. In particular, the singular behavior near the instability point is computed and a detailed comparison with that near a critical point is made. Mode coupling effects, involving the viscous and the heat mode, that determine the singular behavior of the thermal conductivity near the critical point, are also responsible for the singular behavior of the pair correlation function and in particular the density–density correlation function near the instability point. The anomalous density fluctuations could be measured directly by microwave scattering experiments, while the velocity fluctuation could be measured by laser Doppler velocimetry.

KEY WORDS: Kinetic theory; Rayleigh–Bénard cell; mode coupling; correlation functions.

1. INTRODUCTION

In three previous papers the density fluctuations in a fluid in a nonequilibrium stationary state in the presence of a fixed temperature gradient were studied. In particular, the changes in the light scattering from that by a fluid in thermal equilibrium were obtained.[1–4] In all this work the fluid was always in a stable stationary state with no convection present. In this paper we consider the density fluctuations in a fluid in a Rayleigh–Bénard cell near its first convective instability. In that case the fluid is heated from

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below and the presence of a gravitational field and boundary conditions at the wall of the cell have to be explicitly taken into account. The nonconvecting state considered previously is then potentially unstable due to the thermal expansion of the fluid near its lower (heated) boundary.[5] For this nonconvecting state to become unstable the buoyancy force, which is proportional to $\alpha_T g|\nabla T|$, must overcome the dissipative forces, which are proportional to νD_T. Here g is the magnitude of the gravitational acceleration $\mathbf{g}$, $|\nabla T|$ is the absolute value of the temperature gradient ∇T, which is taken to be in the direction of $\mathbf{g}$, $\alpha_T = -(\partial\rho/\partial T)_p/\rho$ is the coefficient of thermal expansion, ρ is the mass density, ν the kinematic viscosity, and D_T is the thermal diffusivity. The dimensionless parameter characterizing these opposing forces is the Rayleigh number $R = \alpha_T g|\nabla T|d^4/\nu D_T$, where d is the distance between the parallel plates that bound the fluid in the direction of the gravitational force and the temperature gradient.

This instability is demonstrated mathematically by showing that one of the eigenvalues, which describes the decay of an arbitrary perturbation of this steady state, goes to zero for some value of R so that then perturbations do not decay. The minimum value of R for which this occurs is known as the critical Rayleigh number and is denoted by R_c. We remark that for $R \geqslant R_c$ there is macroscopic fluid motion which is for the case we consider here, of a fluid only bounded in the direction of $\mathbf{g}$ (and ∇T), typically in the form of two-dimensional rolls.[5]

We have made our calculations in two independent ways: using kinetic theory and hydrodynamic equations. The advantage of the latter is that its results apply to fluids of all densities, while those of the former are restricted to low-density gases. Although we could present our derivations on the basis of hydrodynamic equations, we will use kinetic theory. Not only is this the first time, we believe, that a hydrodynamic instability is treated in the context of kinetic theory, but also some of the connections we want to make are more clearly exposed in such a presentation. In addition, we have explicitly checked that a number of neglected terms are indeed of higher order in the density. Kinetic theory is, therefore, better founded than the more general hydrodynamic theory that is based on rather formal derivations (cf. Refs. 1, 4). We shall use kinetic theory to calculate the long-wavelength part of both the equal and unequal time density–density correlation functions (ddcfs) just below and above the convective instability at $R = R_c$. We will, however, quote our final results in a form valid for arbitrary densities as would follow from a hydrodynamic treatment.

For both $R \lesssim R_c$ and $R \gtrsim R_c$ we shall show that the unequal time correlation functions exhibit critical slowing down similar to that found near a gas–liquid critical point and that in addition, the equal time correlation functions become singular as $|R - R_c| \to 0$. The singular parts of the equal time correlation functions are due to mode-coupling contributions to the nonequilibrium pair correlation function that have been discussed before away from the instability point.[3,4] In fact, in Refs. 3, 4 it was shown that even when the fluid is not close to a hydrodynamic instability similar mode-coupling effects produce a correlation length of the density fluctuations of macroscopic size. Here we show that when R approaches R_c, the length of these correlations in the directions perpendicular to ∇T and $\mathbf{g}$ increases and becomes much larger than d, the size of the cell in the direction of ∇T and $\mathbf{g}$. We remark that in this paper by mode coupling we mean contributions to the pair correlation function that involve the sum of two hydrodynamic eigenvalues or the product of two hydrodynamic eigenfunctions (cf. Sections 4 and 6).

Some of our results—especially for $R \lesssim R_c$—have been found before by others using fluctuating hydrodynamics. The first to compute hydrodynamic fluctuations near the convective instability were Zaitsev and Shliomis.[6] Generalizing the linear fluctuating hydrodynamic equations of Landau and Lifshitz for fluctuations in fluids around total equilibrium to fluids in nonequilibrium steady states, they computed both the equal and unequal time temperature–temperature correlation functions for $R < R_c$. Thus, using fluctuating Navier–Stokes equations linearized about a steady state, they found that these correlation functions exhibit critical slowing down as $R \to R_c$ and that the equal time correlation functions become long ranged. These results are of a similar general form as our Eq. (4.14); however, the correlation length they find differs from ours, given by Eq. (C.8), by a factor of $\sqrt{3}$. The next authors to compute correlation functions near the convective instability were Lesnikov and Fisher.[7] They attempted to compute the equal and unequal time ddcfs for $R < R_c$, i.e., the light scattering, by extending the Onsager regression hypothesis to nonequilibrium steady states for the calculation of the time-dependent correlation functions and then computing the equal time correlation functions by simply taking the equilibrium results with position-dependent thermodynamic parameters. Although, as has been discussed elsewhere,[1-4] the Onsager regression hypothesis can indeed be straightforwardly extended to nonequilibrium steady states, the correct equal time correlation functions cannot be simply obtained from the equilibrium equal time correlation functions. The result of this incorrect extension of equilibrium results is that the contributions of the nonequilibrium part of the pair correlation function to the equal time correlation functions are neglected. As a consequence their equal time correlation functions do not become long ranged as R approaches R_c. Therefore, the criticism of Lesnikov and Fisher on the results of Zaitsev and Shliomis, who did find long-ranged equal time correlation functions, is not justified. Following Lesnikov and Fisher, Lekkerkerker and Boon[8] computed the light scattering, i.e., the ddcfs near the convective instability for $R < R_c$. They also used the Onsager regression hypothesis to compute the time-dependent correlation function and found critical slowing down. They did not, however, specify the equal time ddcf and in addition assumed that the equal time entropy–momentum correlation function is zero. Although the latter is true in equilibrium due to symmetry, in nonequilibrium it is not. As a consequence, the form of their time-dependent correlation functions is not correct, i.e., what corresponds to the second term in Eq. (4.9a) is absent in the results of their calculations, since this term is proportional to the equal time entropy–momentum correlation function.

Finally, Graham,[9] Graham and Pleiner,[10] and Swift and Hohenberg[11] used *non*linear fluctuating hydrodynamics to examine corrections to the results of Zaitsev and Shliomis[6] very close to $R = R_c$. They found that these correlations only become appreciable in an experimentally inaccessible region near R_c. This implied that, unlike near the gas–liquid critical point, fluctuation renormalization of the hydrodynamic eigenvalues (i.e., of the transport coefficients) can be neglected and a linear theory can be used. These points are verified on the basis of kinetic theory in Appendices B and C of this paper. Apart from a detailed discussion of the behavior of density fluctuations for $R \gtrsim R_c$, the main result of this paper that is not evident from previous papers is that essentially the same mode-coupling effects that are responsible for the singular behavior of the thermal conductivity near the gas–liquid critical point are also responsible for the anomalous behavior of the pair correlation function and the ddcfs near the instability point. Although both singularities—those near the critical point and near the instability point—are related to the behavior of the pair correlation function, it is the dynamical nonequilibrium contribution to the pair correlation function, proportional to the gradient of the temperature, that causes the singularity near the instability point, while static contributions, related to the infinite compressibility, lead to the singularity near the critical point.

The quantities we compute are all related to the microscopic density $f(1t)$ in μ space:

$$f(1t) = \sum_{i=1}^{N} \delta(1 - x_i(t)) \tag{1.1}$$

In Eq. (1.1) $1 \equiv (\mathbf{R}_1, \mathbf{V}_1)$ is a particular point in μ space, $x_i(t) \equiv (\mathbf{r}_i(t), \mathbf{v}_i(t))$ is the phase of particle i at time t, and the sum is over the number of particles, N, in a volume Ω.

In the course of our explicit calculations, we will consider fluids that are of finite extent in only one of the three spatial directions and infinite in the remaining two. For this case the limit $N, \Omega \to \infty$, $N/\Omega = n$ is to be used. If we define a density fluctuation in μ space by

$$\delta f(1t) = f(1t) - \langle f(1t) \rangle_{ss} \tag{1.2}$$

where $\langle\ \rangle_{ss}$ denotes a steady state ensemble average, then it is the aim of this paper to calculate for a dilute gas, the unequal time correlation function $C(1t\,|\,2)$

$$C(1t\,|\,2) = \langle \delta f(1t) \delta f(20) \rangle_{ss} \tag{1.3}$$

and the pair correlation function

$$G_2(12) = f_2(12) - f_1(1) f_1(2) \tag{1.4}$$

Here $f_2(12) = \langle \sum_{i \neq j}^{N} \delta(1 - x_i) \delta(2 - x_j) \rangle_{ss}$ is the two-particle distribution function and $f_1(1) = \langle \sum_{i=1}^{N} \delta(1 - x_i) \rangle_{ss}$ is the one-particle distribution function. $C(1t\,|\,2)$ and $G_2(12)$ describe the microscopic correlations in μ space that exist in a gas, in particular also those near $R = R_c$. Further, from these two quantities, the density–density equal and unequal time correlation functions that are typically measured in scattering experiments can be easily determined. For example, one only has to multiply $C(1t\,|\,2)$ by $\int d\mathbf{V}_1 \int d\mathbf{V}_2 m^2$ to obtain $\langle \delta\rho(\mathbf{R}_1, t) \delta\rho(\mathbf{R}_2, 0) \rangle_{ss}$ where m is the mass of particles and $\delta\rho(\mathbf{R}) = \rho(\mathbf{R}) - \langle \rho(\mathbf{R}) \rangle_{ss}$ the density fluctuation at $\mathbf{R}$, with $\rho(\mathbf{R})$ the microscopic mass density at $\mathbf{R}$:

$$\rho(\mathbf{R}) = \sum_{i=1}^{N} m \delta(\mathbf{R} - \mathbf{r}_i) \tag{1.5}$$

The central quantities needed for our calculations are the long-wavelength eigenmodes of a linear kinetic operator, that will be defined in Section 2 [cf. Eqs. (2.4) and (2.17c)]. The relevance of these modes follows from the fact that the eigenvalue which goes to zero near a hydrodynamic instability is a long-wavelength mode. For an equilibrium fluid in infinite space the long-wavelength spectrum or hydrodynamic modes are well known.[12] There are five of these modes: a heat mode (H), two sound modes ($\sigma = \pm 1$), and two viscous modes ($\nu_{1,2}$). Here we compute the nonequilibrium extension of some of these modes for fluids which are in a nonequilibrium steady state in the presence of gravity and of walls.

The plan of this paper is as follows. In the first part of Section 2, we indicate how the equations of Ernst and Cohen,[13] Krommes and Oberman,[14] and Ernst and Dorfman[15] for the unequal and equal time correlation functions can be modified to include both the effects of walls which surround the fluid and a constant gravitational force. The effects of the walls are taken into account using the wall–particle collision operator of Dorfman and van Beijeren.[16] The resulting basic kinetic equations for

$C(1t|2)$ and $G_2(12)$ are linear and involve a linear kinetic operator the eigenvalue spectrum of which is of central interest in this paper. In the second part of Section 2 we derive the equations from which both the right and left hydrodynamic eigenmodes of this operator for gases in a nonequilibrium steady state can be derived. In Section 3 the relevant hydrodynamic modes for $R < R_c$ are obtained explicitly. In Section 4 the results of Section 3 are used to calculate the long-wavelength contributions to both the pair correlation function, $G_2(12)$, and the time-dependent correlation function, $C(1t|2)$, for $R < R_c$. In particular, we concentrate on the contributions to these correlation functions that become singular as R approaches R_c from below. In Section 5 and Appendix A the relevant hydrodynamic modes for $R \gtrsim R_c$ will be obtained explicitly. In Section 6 these modes will be used to calculate the long-wavelength contributions to $G_2(12)$ and $C(1t|2)$ for $R \gtrsim R_c$. In Section 7 the results of this paper are reviewed. In particular the singular behavior found here near the hydrodynamic instability is compared with that found near the critical point for the gas–liquid phase transition. Also some remarks about experimental verification of some of the new results are made. In Appendix B we shall show that as R approaches R_c thermodynamic fluxes and transport coefficients, in particular the heat flux and the thermal conductivity, become renormalized and singular due to correlated collision sequences (ring events) between the particles. In Appendix C the consequences of this on the theory presented in this paper are shown to be negligible.

Some of the main results quoted in this paper have been published in a short previous publication.[17]

2. BASIC EQUATIONS AND HYDRODYNAMIC EIGENMODES

In this section we first give the basic equations for the unequal and equal time correlation functions in μ space that determine the corresponding correlation functions, in particular the density–density correlation functions, in ordinary space. As mentioned in Section 1, in these equations a basic linear kinetic operator occurs, certain eigenmodes of which are of particular importance for our calculations. The second part of this section is therefore devoted to developing a general method to determine the hydrodynamic modes of this operator for a gas in a nonequilibrium steady state in a finite container. In the following sections we shall use this method to determine approximate hydrodynamic modes below and above the convective instability and use them to calculate the singular behavior of the correlation functions introduced in the first part of this section near the instability point $R = R_c$.

2.1. Basic Equations

On the basis of the methods developed by Ernst and Cohen,[13] Krommes and Oberman,[14] Ernst and Dorfman,[15] and Dorfman and van Beijeren[16] kinetic equations for the correlation functions of a dilute gas in a nonequilibrium steady state in the presence of walls and external forces can be derived without difficulty.[18] The unequal time correlation function $C(1t\,|\,2)$ [cf. Eq. (1.3)] satisfies for $t > 0$, the equation

$$\left[\partial_t + L_{ss}(1)\right]C(1t\,|\,2) = \overline{T}_w(1)C(1t\,|\,2) \tag{2.1a}$$

Equation (2.1a) is to be solved in terms of the equal time correlation function:

$$C(1t=0\,|\,2) \equiv C(1\,|\,2) = \delta(1-2)f_1(1) + G_2(12) \tag{2.1b}$$

In Eq. (2.1b) $f_1(1)$ is defined below Eq. (1.4) and $G_2(12)$ by Eq. (1.4). For dilute gases in a steady state in the presence of gravity and walls $f_1(1)$ satisfies the extended nonlinear Boltzmann equation[16]:

$$\left\{\mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1} + \mathbf{g}\cdot\frac{\partial}{\partial\mathbf{V}_1}\right\}f_1(1) = \int d2\,\hat{T}(12)f_1(1)f_1(2) + \overline{T}_w(1)f_1(1) \tag{2.1c}$$

where $G_2(12)$ satisfies the equation

$$\left[L_{ss}(1) + L_{ss}(2) - \overline{T}_w(1) - \overline{T}_w(2)\right]G_2(12) = \hat{T}(12)f_1(1)f_1(2) \tag{2.1d}$$

In Eq. (2.1) $L_{ss}(1)$ is a linear kinetic operator, obtained by linearizing the nonlinear operator appearing in Eq. (2.1c) around f_1:

$$L_{ss}(1) = \mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1} + \mathbf{g}\cdot\frac{\partial}{\partial\mathbf{V}_1} - \int d3\,\hat{T}(13)(1+P_{13})f_1(3) \tag{2.2a}$$

with $\hat{T}(13)$ the point binary collision operator[13]:

$$\hat{T}(13) = \delta(\mathbf{R}_1-\mathbf{R}_3)T_0(13) = \delta(\mathbf{R}_1-\mathbf{R}_3)\int_0^{2\pi}d\epsilon\int_0^a db\,b|\mathbf{V}_1-\mathbf{V}_3|(b_\sigma - 1) \tag{2.2b}$$

where (b,ϵ) are, respectively, the impact parameter and azimuthal angle of the binary collision between two particles with velocities $\mathbf{V}_1, \mathbf{V}_3$, a is the range of the interparticle forces, and b_σ is an operator that replaces the velocities $\mathbf{V}_1$ and $\mathbf{V}_3$ by the velocities of the restituting collision, $\mathbf{V}_1'$ and $\mathbf{V}_3'$. Further, P_{13} in Eq. (2.2a) is a permutation operator that permutes particle indices 1 and 3. $\overline{T}_w(1)$ in Eq. (2.1) is a wall–particle collision operator that takes into account the change in the distribution functions due to collisions of the particles with the walls. Under quite general conditions it has been shown[16] that when $\overline{T}_w(1)$ acts on a function $h(1)$ the result is given by

$$\overline{T}_w(1)h(1) = \int dw\,\delta(\mathbf{R}_1 - \mathbf{R}_w)$$
$$\times \left\{ \theta(\mathbf{V}_1 \cdot \hat{n}) \int d\mathbf{V}_1' \,\theta(-\mathbf{V}_1' \cdot \hat{n})\, |V_1' \cdot \hat{n}| K(\mathbf{V}_1, \mathbf{V}_1') h(\mathbf{R}_1, \mathbf{V}_1') - \theta(-\mathbf{V}_1 \cdot \hat{n}) |\mathbf{V}_1 \cdot \hat{n}| h(1) \right\} \tag{2.2c}$$

where $\mathbf{R}_w$ denotes the position of a point on the walls, $\int dw$ indicates an integral over the wall surfaces, $\theta(x) = 1$ for $x > 0$ and is zero otherwise, $\hat{n}$ is a unit vector normal to the wall pointing into the fluid, and $K(\mathbf{V}_1, \mathbf{V}_1')$ is a scattering kernel which specifies the interaction mechanism between the walls and the gas particles. An explicit example of $K(\mathbf{V}_1, \mathbf{V}_1')$ will be given below.

To determine the long-wavelength contributions to the correlation functions of the fluctuations, the average state, around which the properties of the fluctuations are studied, i.e., f_1, is needed first. For this will determine the operator $L_{ss}(1)$, Eq. (2.2a), that occurs in Eqs. (2.1a) and (2.1d) for $C(1t)$ and $G_2(12)$, respectively, as well as the right-hand side of Eq. (2.1d). We remark that, unlike in equilibrium, $G_2(12)$ possesses nonequilibrium contributions that are of the same order in the density as $f_1(1)$, so that $G_2(12)$ cannot be neglected in Eqs. (2.1b) and (2.1d).[1-4] We proceed now as follows.

First the Chapman–Enskog solution method is used to solve Eq. (2.1c) for $f_1(1)$, which is then obtained as an expansion in terms of a uniformity parameter $\mu = l/L_\nabla$ with l the mean free path of the particles between successive collisions and L_∇ a macroscopic gradient length on the order of $T/|\nabla T|$.[3] Using that $f_1(1)$ vanishes outside the fluid volume, the first two terms in this expansion are straightforwardly found to be[16]

$$f_1(1) \equiv f_1^{\text{CH-E}}(1) = W(\mathbf{R}_1)\left[f_l(1) + f_\nabla^{(1)}(1) + O(\mu^2) \right] \tag{2.3a}$$

Here $W(\mathbf{R}_1)$ is a characteristic function that vanishes when $\mathbf{R}_1$ is outside the fluid volume Ω:

$$\begin{aligned} W(\mathbf{R}_1) &= 1 \qquad \text{if} \quad \mathbf{R}_1 \in \Omega \\ &= 0 \qquad \text{otherwise} \end{aligned} \tag{2.3b}$$

In Eq. (2.3a) $f_l(1)$ is the local Maxwellian distribution function given by

$$f_l(1) = n(\mathbf{R}_1)\left[\frac{m}{2\pi k_B T(\mathbf{R}_1)} \right]^{3/2} \exp - \left[\frac{m C_1^2(\mathbf{R}_1)}{2 k_B T(\mathbf{R}_1)} \right] \tag{2.3c}$$

[3] For a gas at STP, $l \sim 10^{-5}$ cm and $T \sim 300$ K. Using this and that a typical value of $|\nabla T|$ in a Bénard cell is $|\nabla T| \sim 50$ K/cm yields $\mu \sim 10^{-6}$.

where $n(\mathbf{R}_1)$ is the local number density, k_B is Boltzmann's constant, m is the mass of the particles, and $\mathbf{C}_1(\mathbf{R}_1) = \mathbf{V}_1 - \mathbf{u}(\mathbf{R}_1)$ is the peculiar velocity with $\mathbf{u}(\mathbf{R}_1)$ the local flow velocity at the point $\mathbf{R}_1$. Further, $f_\nabla^{(1)}(1)$ is the correction of first order in the gradients to $f_l(1)$ and is given by[16]

$$f_\nabla^{(1)}(1) = \frac{1}{\bar{\Lambda}_l(1)} f_l(1)\,\beta m\left(C_{1\alpha}C_{1\beta} - \frac{\delta_{\alpha\beta}}{3}C_1^2\right)\frac{\partial u_\alpha}{\partial R_{1\beta}} + \frac{1}{\bar{\Lambda}_l(1)} f_l(1)\left(\frac{\beta m C_1^2}{2} - \frac{5}{2}\right)C_{1\alpha}\frac{\partial \log T}{\partial R_{1\alpha}} \tag{2.3d}$$

where $\beta = 1/k_B T$ and in Eq. (2.3d), as in the rest of this paper, summation convention is used. $\bar{\Lambda}_l(1)$ is the collision operator linearized around local equilibrium:

$$\bar{\Lambda}_l(1) = \int d3\,\hat{T}(13)(1 + P_{13})f_l(3) \tag{2.3e}$$

Equation (2.3) determines the average state f_l of the fluid, around which the density fluctuations are studied.

2.2. Hydrodynamic Eigenmodes

The next step in determining the long-wavelength part of $C(1t\,|\,2)$ and $G_2(12)$ is to compute the eigenvalue spectrum of the operator $L_{ss}(1) - \bar{T}_w(1)$ [cf. Eqs. (2.1a) and (2.1d)] for slowly varying disturbances. The right eigenvalue problem is defined as

$$\left[L_{ss}(1) - \bar{T}_w(1)\right] f_l(1)\Theta_j^R(1) = \omega_j f_l(1)\Theta_j^R(1) \tag{2.4}$$

where $\Theta_j^R(1)$ is the right eigenfunction, ω_j is the eigenvalue, j is a general eigenfunction index, and the fact $f_l(1)$ has been inserted for convenience. Before defining the adjoint or left eigenvalue problem, we continue with a discussion of how to calculate $\Theta_j^R(1)$.

Following Dorfman and van Beijeren,[16] we note that the right eigenfunctions of the operator $L_{ss}(1) - \bar{T}_w(1)$ vanish when $\mathbf{R}_1$ is not inside the fluid volume.[4] Using this, we define

$$\Theta_j^R(1) = W(\mathbf{R}_1)\tilde{\Theta}_j^R(1) \tag{2.5}$$

[4] Since these eigenfunctions will be used to calculate, for example, the pair correlation function $G_2(12)$ and since $G_2(12)$ vanishes outside the fluid volume, it follows that for $\mathbf{R}_i \notin \Omega$, $\Theta_j^R(\mathbf{R}_i, \mathbf{V}_i)$ must also vanish [cf. Eq. (4.1)].

Inserting Eq. (2.5) into Eq. (2.4) yields

$$\begin{aligned} W(\mathbf{R}_1)\left[L_{ss}(1)-\omega_j\right]f_l(1)\tilde{\Theta}_j^R(1) &= \bar{T}_w(1)f_l(1)\tilde{\Theta}_j^R(1) - f_l(1)\tilde{\Theta}_j^R(1)\mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1}W(\mathbf{R}_1) \\ &\equiv T_w(1)f_l(1)\tilde{\Theta}_j^R(1) \end{aligned} \tag{2.6a}$$

where

$$\begin{aligned} T_w(1)f_l(1)\tilde{\Theta}_j^R(1) = \int dw\,\theta(\mathbf{V}_1\cdot\hat{n})\delta(\mathbf{R}_1-\mathbf{R}_w) &\times\left\{\int d\mathbf{V}_1'\,\theta(-\mathbf{V}_1'\cdot\hat{n})|\mathbf{V}_1'\cdot n|K(\mathbf{V}_1,\mathbf{V}_1')f_l(\mathbf{R}_1,\mathbf{V}_1')\right. \\ &\left.\times\,\tilde{\Theta}_j^R(\mathbf{R}_1,\mathbf{V}_1') - |\mathbf{V}_1\cdot n|f_l(1)\tilde{\Theta}_j^R(1)\right\} \end{aligned} \tag{2.6b}$$

If we now assume that $\tilde{\Theta}_j^R(1)$ is continuous at the surfaces of the container, it then follows that each side of Eq. (2.6a) must vanish separately since the right-hand side of this equation vanishes everywhere except at the walls where it is discontinuous. Physically, this is equivalent to requiring that there are no sources or sinks of particles at the wall.[(16)] The resulting two equations are

$$L_{ss}(1)f_l(1)\tilde{\Theta}_j^R(1) = \omega_j f_l(1)\tilde{\Theta}_j^R(1) \tag{2.6c}$$

and

$$T_w(1)f_l(1)\tilde{\Theta}_j^R(1) = 0 \tag{2.6d}$$

For $\mathbf{V}_1\cdot\hat{n} > 0$, Eq. (2.6d) is equivalent to

$$\begin{aligned} &|\mathbf{V}_1\cdot\hat{n}|f_l(\mathbf{R}_w,\mathbf{V}_1)\tilde{\Theta}_j^R(\mathbf{R}_w,\mathbf{V}_1) \\ &\quad = \int d\mathbf{V}_1'\,\theta(-\mathbf{V}_1'\cdot\hat{n})|\mathbf{V}_1'\cdot\hat{n}|K(\mathbf{V}_1,\mathbf{V}_1')f_l(\mathbf{R}_w,\mathbf{V}_1')\tilde{\Theta}_j^R(\mathbf{R}_w,\mathbf{V}_1') \end{aligned} \tag{2.6e}$$

Equation (2.6e) will be used to determine boundary conditions on the right eigenfunctions $\tilde{\Theta}_j^R(1)$. Next we show that Eqs. (2.6c) and (2.6e) are equivalent to a system of differential equations with boundary conditions for the unknown $\tilde{\Theta}_j^R(1)$ and ω_j.

In order to derive this, we use that we are interested in slowly varying disturbances in space and time where the parameter $l/d \ll 1$. Here d is the distance over which $\tilde{\Theta}_j^R$ changes, e.g., in the Bénard problem, the distance between the parallel plates that bound the fluid.[5] Since [cf. (2.2a)] the

[5] Typically, $d \cong 0.1$ cm.

collision operator $\int d3\,\hat{T}(13)(1 + P_{13})$ in Eq. (2.6c) is of $O(l^{-1})$ and $\partial/\partial\mathbf{R}_1$ in this equation is of $O(d^{-1})$, we see that expansion parameter l/d will appear naturally in the solution of Eq. (2.6c).

We make use of the fact that we are interested in slowly varying disturbances in time in treating also the eigenvalue, ω_j, in Eq. (2.6c) as small.[6] For the long-time behavior we are interested in here, only the smallest eigenvalues related to the conserved quantities in a binary collision are needed then. Thus, we introduce a kinetic (local equilibrium) projection operator that can be symbolically written in the convenient compact form

$$P(1) = \sum_{i=1}^{5} \frac{|\psi_i(1)\rangle_l\langle\psi_i(1)|}{\langle\psi_i(1)\,|\,\psi_i(1)\rangle_l} \tag{2.7a}$$

$P(1)$ projects functions onto the space of the five conserved quantities in a binary collision: the $\psi_i(1)$ $(i = 1, 2, \ldots, 5)$ given by

$$\psi_1(1) = m, \qquad \psi_{i=2,3,4}(1) = mC_{1x}(\mathbf{R}_1), \quad mC_{1y}(\mathbf{R}_1), \quad mC_{1z}(\mathbf{R}_1)$$
$$\psi_5(1) = \left(\beta(\mathbf{R}_1)\frac{mC_1^2(\mathbf{R}_1)}{2} - \frac{3}{2}\right) \tag{2.7b}$$

In Eq. (2.7a) we have defined an inner product in velocity space for two arbitrary functions f and g by

$$\langle f(1)\,|\,g(1)\rangle_l \equiv \int d\mathbf{V}_1\, f(1)g(1)\phi_l(1) \tag{2.7c}$$

with $\phi_l(1) = f_l(1)/n(\mathbf{R}_1)$. The symbol $|\ \rangle_l$ denotes that the weight function ϕ_l is associated with the ket vector. In general, the bra–ket notation $\langle\psi_i(1)| \equiv \psi_i(1)$ and $|\psi_i\rangle_l \equiv \psi_i(1)\phi_l(1)$ is used only for convenience and mainly to obtain a compact notation for inner products, as defined by the bracket in Eq. (2.7c).

Using Eq. (2.7), we write $f_l(1)\tilde{\Theta}_j^R(1)$ as

$$f_l(1)\tilde{\Theta}_j^R(1) = P(1)f_l(1)\tilde{\Theta}_j^R(1) + P_\perp(1)f_l(1)\tilde{\Theta}_j^R(1) \tag{2.8}$$

where $P_\perp(1) = 1 - P(1)$.

From Eqs. (2.8) and (2.6c) equations for $P(1)f_l(1)\tilde{\Theta}_j^R(1)$ and $P_\perp(1)f_l(1)\tilde{\Theta}_j^R(1)$ can be constructed which, when solved, will yield $f_l(1)\tilde{\Theta}_j^R(1)$ by Eq. (2.8). Thus, multiplying Eq. (2.6c) by $P(1)$ and $P_\perp(1)$, respectively, using Eq. (2.8), solving the equation for $P_\perp(1)f_l(1)\tilde{\Theta}_j^R(1)$, and then inserting the

[6] Since ω_j has the units of inverse time, we actually use that ω_j is much less than $t_0^{-1} = V_{\text{th}}/l$ where V_{th} is the thermal velocity.

result into the equation for $P(1)f_l(1)\tilde{\Theta}_j^R(1)$ yields the following two equations for $P_\perp(1)f_l(1)\tilde{\Theta}_j^R(1)$ and $P(1)f_l(1)\tilde{\Theta}_j^R$, respectively:

$$P_\perp(1)f_l(1)\tilde{\Theta}_j^R(1) = -P_\perp(1)\frac{1}{[P_\perp(1)L_{ss}(1)P_\perp(1)-\omega_j]}L_{ss}(1)P(1)f_l(1)\tilde{\Theta}_j^R(1) \tag{2.9a}$$

and

$$\left\{P(1)L_{ss}(1)P(1) - P(1)L_{ss}(1)P_\perp(1)\frac{1}{[P_\perp(1)L_{ss}(1)P_\perp(1)-\omega_j]}\times P_\perp(1)L_{ss}(1)P(1)\right\}P(1)f_l(1)\tilde{\Theta}_j^R(1) = \omega_j P(1)f_l(1)\tilde{\Theta}_j^R(1) \tag{2.9b}$$

We remark that Eqs. (2.9) and (2.8) are formally exact equations for $f_l(1)\tilde{\Theta}_j^R(1)$.

In order to put these formal equations into a manageable form, we make use of the three small parameters that appear in the theory: (1) $l/L_\nabla \ll 1$, (2) $l/d \ll 1$, and (3) $\omega_j t_0 \ll 1$, where t_0 is the mean free time.[6] Then Eq. (2.9b) can be written to first order in these small parameters:

$$\left\{P(1)\left[\mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1} + \mathbf{g}\cdot\frac{\partial}{\partial\mathbf{V}_1}\right]P(1) + P(1)\left[\mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1} + \mathbf{g}\cdot\frac{\partial}{\partial\mathbf{V}_1}\right]P_\perp(1)\frac{1}{\overline{\Lambda}_l(1)}P_\perp(1) \times\left[\mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1} + \mathbf{g}\cdot\frac{\partial}{\partial\mathbf{V}_1} - \overline{\Lambda}_\nabla^{(1)}(1)\right]P(1)\right\}P(1)f_l(1)\tilde{\Theta}_j^R(1) = \omega_j P(1)f_l(1)\tilde{\Theta}_j^R(1) \tag{2.10a}$$

where

$$\overline{\Lambda}_\nabla^{(1)}(1) \equiv \int d3\,\hat{T}(13)(1+P_{13})f_\nabla^{(1)}(3) \tag{2.10b}$$

Equations (2.10) represent five coupled hydrodynamiclike equations that can be used to calculate $f_l(1)\tilde{\Theta}_j^R(1)$ and ω_j to lowest order in μ, i.e., to $\mathcal{O}(\mu^0)$.[7]

The explicit form of these equations can be determined as follows. We first define the hydrodynamic part of the right eigenfunction $\tilde{\Theta}_j^R(1)$ by the

[7] We do not give the equation for $P_\perp(1)f_l(1)\tilde{\Theta}_j^R(1)$ since it is of $O(\mu)\cdot[P(1)\tilde{\Theta}_j^R(1)]$ and not needed in our calculations.

equation

$$
\begin{aligned}
&P(1)f_l(1)\tilde{\Theta}_j^R(1) \\
&\quad = \sum_{i=1}^{5} |\psi_i(1)\rangle_l \frac{\langle\psi_i(1)n(\mathbf{R}_1)\tilde{\Theta}_j^R(1)\rangle_l}{\langle\psi_i(1)\,|\,\psi_i(1)\rangle_l} \\
&\quad = \frac{|\psi_1\rangle_l}{m^2}\hat{\rho}_j(\mathbf{R}_1) + \frac{\beta}{m}|\psi_\nu\rangle_l[\hat{p}_{j\nu}(\mathbf{R}_1) - u_\nu\hat{\rho}_j(\mathbf{R}_1)] + |\psi_5\rangle_l n\frac{\hat{T}_j(\mathbf{R}_1)}{T}
\end{aligned}
\tag{2.11}
$$

Equation (2.11) can be regarded as a definition of the first few velocity moments of $\tilde{\Theta}_j^R(1)$. Inserting Eq. (2.11) into Eq. (2.10) and multiplying the resulting equation by $\int d\mathbf{V}_1\psi_i(1)$ $(i = 1, \ldots, 5)$, yields five coupled hydrodynamiclike equations for the unknowns $\hat{\rho}_j$, $\hat{p}_{j\nu}$, $\hat{T}_j$, and ω_j, i.e., a hydrodynamic eigenvalue problem. Using that

$$
\eta_B(\mathbf{R}_1)\Delta_{\alpha\beta,\gamma\nu} = -\frac{m^2 n(\mathbf{R}_1)}{k_B T(\mathbf{R}_1)}\int d\mathbf{V}_1\, C_{1\alpha}C_{1\beta}\frac{1}{\overline{\Lambda}_l(1)}\phi_l(1)\left(C_{1\gamma}C_{1\nu} - \frac{\delta_{\gamma\nu}C_1^2}{3}\right)
\tag{2.12a}
$$

and

$$
\begin{aligned}
\lambda_B(\mathbf{R}_1)\delta_{\alpha\beta} = {}& -k_B n(\mathbf{R}_1)\int d\mathbf{V}_1\, C_{1\alpha}\left(\frac{\beta m C_1^2}{2} - \frac{5}{2}\right)\frac{1}{\overline{\Lambda}_l(1)}\phi_l(1) \\
&\times C_{1\beta}\left(\frac{\beta m C_1^2}{2} - \frac{5}{2}\right)
\end{aligned}
\tag{2.12b}
$$

where $\Delta_{\alpha\beta,\gamma\nu} = [\delta_{\alpha\gamma}\delta_{\beta\nu} + \delta_{\alpha\nu}\delta_{\beta\gamma} - \frac{2}{3}\delta_{\alpha\beta}\delta_{\gamma\nu}]$ and $\eta_B(\lambda_B)$ is the low-density (Boltzmann) value of the shear viscosity (heat conductivity) and performing straightforward but lengthy algebraic manipulations, the resulting explicit equations are found to be

$$
\omega_j\hat{\rho}_j(\mathbf{R}_1) = \frac{\partial}{\partial R_{1\nu}}\hat{p}_{j\nu}(\mathbf{R}_1)
\tag{2.13a}
$$

and

$$
\begin{aligned}
-\omega_j\hat{p}_{j\alpha}(\mathbf{R}_1) = {}& -\frac{\partial}{\partial R_{1\beta}}\Big\{nk_B\hat{T}_j(\mathbf{R}_1)\delta_{\alpha\beta} + \frac{k_B T}{m}\hat{\rho}_j(\mathbf{R}_1)\delta_{\alpha\beta} + u_\beta\hat{p}_{j\alpha}(\mathbf{R}_1) \\
&\quad + u_\alpha\hat{p}_{j\beta}(\mathbf{R}_1) - u_\alpha u_\beta\hat{\rho}_j(\mathbf{R}_1)\Big\} + g_\alpha\hat{\rho}_j(\mathbf{R}_1) \\
&+ \frac{\partial}{\partial R_{1\beta}}\eta_B\Delta_{\alpha\beta,\gamma\nu}\frac{\partial}{\partial R_{1\gamma}}\frac{[\hat{p}_{j\nu}(\mathbf{R}_1) - u_\nu\hat{\rho}_j(\mathbf{R}_1)]}{\rho} \\
&+ \frac{\partial}{\partial R_{1\beta}}\left(\frac{\partial\eta_B}{\partial T}\right)_\rho\hat{T}_j(\mathbf{R}_1)2\left[D_{\alpha\beta} - \frac{\delta_{\alpha\beta}}{3}D_{\gamma\gamma}\right]
\end{aligned}
\tag{2.13b}
$$

and

$$-\omega_j \frac{3}{2} nk_B \hat{T}_j(\mathbf{R}_1) = -\frac{3}{2} nk_B u_\nu \frac{\partial}{\partial R_{1\nu}} \hat{T}_j(\mathbf{R}_1) - \frac{3}{2} nk_B \frac{\hat{p}_{j\nu}(\mathbf{R}_1)}{\rho} \frac{\partial T}{\partial R_{1\nu}}$$
$$- nk_B T \frac{\partial}{\partial R_{1\nu}} \frac{\left[\hat{p}_{j\nu}(\mathbf{R}_1) - u_\nu \hat{\rho}_j(\mathbf{R}_1)\right]}{\rho}$$
$$+ \frac{\partial}{\partial R_{1\beta}} \lambda_B \frac{\partial}{\partial R_{1\beta}} \hat{T}_j(\mathbf{R}_1) + \frac{\partial}{\partial R_{1\beta}} \left(\frac{\partial \lambda_B}{\partial T}\right)_\rho \hat{T}_j(\mathbf{R}_1) \frac{\partial T}{\partial R_{1\beta}}$$
$$+ 4\eta_B \left[D_{\alpha\beta} - \frac{\delta_{\alpha\beta}}{3} D_{\gamma\gamma}\right] \frac{\partial}{\partial R_{1\alpha}} \frac{\left[\hat{p}_{j\nu}(\mathbf{R}_1) - u_\nu \hat{\rho}_j(\mathbf{R}_1)\right]}{\rho}$$
$$+ 2D_{\alpha\beta} \left(\frac{\partial \eta_B}{\partial T}\right)_\rho \hat{T}_j(\mathbf{R}_1) \left[D_{\alpha\beta} - \frac{\delta_{\alpha\beta}}{3} D_{\gamma\gamma}\right] \tag{2.13c}$$

In Eqs. (2.13) $\rho = mn$ is the local mass density, all hydrodynamic quantities are to be evaluated at the point $\mathbf{R}_1$, and $D_{\alpha\beta} = (1/2)(\partial u_\alpha / \partial R_{1\beta} + \partial u_\beta / \partial R_{1\alpha})$. We remark that Eqs. (2.13) have the same form as the hydrodynamic eigenvalue problem for dilute gases obtained by linearizing the Navier–Stokes equations around a nonequilibrium steady state.

In order to obtain boundary conditions for these differential equations, we use Eq. (2.6e). Examining Eq. (2.9a) and giving $[\bar{\Lambda}_l(1)]^{-1}$ the weight l, we see by expansion around $\bar{\Lambda}_l(1)^{-1}$ that $P_\perp(1) f_l(1) \tilde{\Theta}_j^R(1) \sim O(\mu) \cdot [P(1) f_l(1) \tilde{\Theta}_j^R(1)]$. Requiring Eq. (2.6e) to be satisfied to each order in μ and using Eq. (2.11) yields

$$|\mathbf{V}_1 \cdot \hat{n}| \phi_l(\mathbf{R}_w, \mathbf{V}_1) \left\{ \frac{\hat{\rho}_j(\mathbf{R}_w)}{m^2} + \beta(\mathbf{R}_w) C_{1\nu}(\mathbf{R}_{1w}) \left[\hat{p}_{j\nu}(\mathbf{R}_w) - u_\nu(\mathbf{R}_w) \hat{\rho}_j(\mathbf{R}_w)\right] \right.$$
$$\left. + \left[\frac{\beta(\mathbf{R}_w) m C_1^2(\mathbf{R}_w)}{2} - \frac{3}{2}\right] n(\mathbf{R}_w) \frac{\hat{T}_j(\mathbf{R}_w)}{T(\mathbf{R}_w)} \right\}$$
$$= \int d\mathbf{V}_1' \, \theta(-V_1' \cdot \hat{n}) |\mathbf{V}_1' \cdot \hat{n}| K(\mathbf{V}_1, \mathbf{V}_1') \phi_l(\mathbf{R}_w, \mathbf{V}_1')$$
$$\times \left\{ \frac{\hat{\rho}_j(\mathbf{R}_w)}{m^2} + \beta(\mathbf{R}_w) C_{1\nu}'(\mathbf{R}_w) \left[\hat{p}_{j\nu}(\mathbf{R}_w) - u_\nu(\mathbf{R}_w) \hat{\rho}_j(\mathbf{R}_w)\right] \right.$$
$$\left. + \left[\frac{\beta(\mathbf{R}_w) m C_1'^2(\mathbf{R}_w)}{2} - \frac{3}{2}\right] n(\mathbf{R}_w) \frac{\hat{T}_j(\mathbf{R}_w)}{T(\mathbf{R}_w)} \right\} \tag{2.14}$$

From Eq. (2.14) boundary conditions on $\hat{\rho}_j$, $\hat{p}_{j\nu}$, and $\hat{T}_j$ to $O(\mu^0)$ can be determined once $K(\mathbf{V}_1, \mathbf{V}_1')$ is given. To illustrate the derivation of boundary conditions, we assume diffusive reflection of the particles at the wall which implies that a molecule striking the wall is absorbed and instantly reemitted with a velocity determined by a Maxwell distribution with the temperature $T_w = (k_B \beta_w)^{-1}$ of the wall. For this case $K(\mathbf{V}_1, \mathbf{V}_1') \equiv K_D(\mathbf{V}_1, \mathbf{V}_1')$ is given by[16]

$$K_D(\mathbf{V}_1, \mathbf{V}_1') = |\mathbf{V}_1 \cdot \hat{n}|(2\pi m \beta_w)^{1/2} \left(\frac{m\beta_w}{2\pi} \right)^{3/2} \exp\left(\frac{-mV_1^2}{2k_B T_w} \right) \qquad (2.15a)$$

Using that, for this scattering kernel, the boundary conditions on $\mathbf{u}$ and T to $O(\mu^0)$ for stationary walls are[16] $\mathbf{u}(\mathbf{R}_w) = 0$ and $T(\mathbf{R}_w) = T_w$ and using Eq. (2.15a) in Eq. (2.14) yields

$$\begin{aligned} \hat{p}_{j\nu}(\mathbf{R}_w) &= 0 \\ \hat{T}_j(\mathbf{R}_w) &= 0 \end{aligned} \qquad (2.15b)$$

Equations (2.8), (2.10), (2.11), (2.13), and (2.15b) form a closed set of equations by which the right eigenmodes can be determined to $O(\mu)$.[8]

To complete the calculation of the nonequilibrium hydrodynamic modes, we need to determine equations for the adjoint or left eigenfunctions $\Theta_j^L(1)$. This will be done in a manner analogous to that just given for Θ_j^R. To proceed, we normalize the eigenfunctions by requiring

$$\left(\Theta_k^L(1) f_l(1) \Theta_j^R(1)\right) \equiv \int d\mathbf{R}_1 \int d\mathbf{V}_1\, \Theta_k^L(1) f_l(1) \Theta_j^R(1) = \delta_{jk} \qquad (2.16)$$

where the integration over $\mathbf{R}_1$ is over all space and δ_{jk} is in general a product of Dirac and Kronecker delta functions depending on whether the set of indices j and k are continuous or discrete. In Eq. (2.16), we have defined the brackets ( ) to be an inner product in both velocity and position space with no weight function. Further, because $\Theta_j^R(1)$ in Eq. (2.16) is proportional to the characteristic function $W(\mathbf{R}_1)$ [cf. Eq. (2.3b)], the spatial integral in Eq. (2.16) is actually restricted to the fluid volume Ω. To determine the kinetic equation for $\Theta_k^L(1)$, we multiply Eq. (2.4) by $(\Theta_k^L(1)|$, use Eq. (2.16), and integrate by parts to obtain

$$\begin{aligned} -\Big(f_l(1)\Theta_j^R(1)\Big[\mathbf{V}_1 \cdot \partial/\partial \mathbf{R}_1 + \mathbf{g} \cdot \partial/\partial \mathbf{V}_1 + \overline{T}_w^+(1) \\ + \int d3\, f_1(3)\hat{T}(13)(1 + P_{13})\Big]\Theta_k^L(1)\Big) \\ = \omega_j\big(f_l(1)\Theta_j^R(1)\Theta_k^L(1)\big) = \omega_k \delta_{jk} \end{aligned} \qquad (2.17a)$$

[8] In our explicit calculations, we compute the eigenvalues to $O(\mu)$ but the eigenfunctions to $O(\mu^0)$ only. Because of this, we need not consider corrections of $O(\mu)$ to the boundary conditions.

where in giving Eq. (2.17a) we have let $\hat{T}(13)(1 + P_{13})$ act backwards and defined the adjoint of $\bar{T}_w(1)$ to be $\bar{T}_w^+(1)$. It is straightforward to construct $\bar{T}_w^+(1)$ from $\bar{T}_w(1)$ with the result

$$\bar{T}_w^+(1)h(1) = \int dw\, \delta(\mathbf{R}_1 - \mathbf{R}_w)\theta(-\mathbf{V}_1 \cdot \hat{n}) \times \left\{ |\mathbf{V}_1 \cdot \hat{n}| \int d\mathbf{V}_1'\, \theta(\mathbf{V}_1' \cdot \hat{n}) K(\mathbf{V}_1', \mathbf{V}_1) h(\mathbf{R}_1, \mathbf{V}_1') - |\mathbf{V}_1 \cdot \hat{n}|\, h(1) \right\} \tag{2.17b}$$

From Eq. (2.17a) it follows that the adjoint kinetic eigenvalue equation is

$$-\left\{ \mathbf{V}_1 \cdot \frac{\partial}{\partial \mathbf{R}_1} + \mathbf{g} \cdot \frac{\partial}{\partial \mathbf{V}_1} + \bar{T}_w^+(1) + \int d3\, f_1(3) \hat{T}(13)(1 + P_{13}) \right\} \Theta_k^L = \omega_k \Theta_k^L(1) \tag{2.17c}$$

From Eq. (2.17c) we shall now obtain a set of hydrodynamic equations that determine $\Theta_k^L(1)$.

Taking $\Theta_k^L(1)$ to be continuous at the walls, we obtain for $\Theta_k^L(1)$ the two equations

$$-\left[\mathbf{V}_1 \cdot \frac{\partial}{\partial \mathbf{R}_1} + \mathbf{g} \cdot \frac{\partial}{\partial \mathbf{V}_1} + \int d3\, f_1(3) \hat{T}(13)(1 + P_{13}) \right] \Theta_k^L(1) = \omega_k \Theta_k^L(1) \tag{2.18a}$$

and

$$\bar{T}_w^+(1)\Theta_k^L(1) = 0 \tag{2.18b}$$

For $\mathbf{V}_1 \cdot \hat{n} < 0$, Eq. (2.18b) is equivalent to

$$\Theta_k^L(\mathbf{R}_w, \mathbf{V}_1) = \int d\mathbf{V}_1'\, \theta(\mathbf{V}_1' \cdot \hat{n}) K(\mathbf{V}_1', \mathbf{V}_1) \Theta_k^L(\mathbf{R}_w, \mathbf{V}_1') \tag{2.18c}$$

From Eq. (2.18c) we will construct boundary conditions for the $\Theta_k^L(1)$.

Hydrodynamic equations and boundary conditions for Θ_k^L can be obtained from Eqs. (2.18a) and (2.18c) in a manner analogous to that given for $\tilde{\Theta}_j^R$. Thus we introduce an adjoint kinetic projection operator:

$$P^+(1)\Theta_k^L(1) \equiv \sum_{i=1}^{5} \frac{|\psi_i(1)\rangle\langle\psi_i(1) | \Theta_k^L(1)\rangle_l}{\langle\psi_i(1) | \psi_i(1)\rangle_l} \equiv |\psi_1\rangle \frac{\hat{\rho}_k^+(\mathbf{R}_1)}{\rho m} + \frac{\beta}{\rho} |\psi_\nu(1)\rangle \left[\hat{p}_{k\nu}^+(\mathbf{R}_1) - u_\nu \hat{\rho}_k^+(\mathbf{R}_1) \right] + |\psi_5(1)\rangle \frac{\hat{T}_k^+(\mathbf{R}_1)}{T} \tag{2.19}$$

Using arguments identical to those given for the $\tilde{\Theta}_j^R$, we can construct equations for $\hat{\rho}^+$, $\hat{p}_\nu^+$, and $\hat{T}^+$ that are valid when $l/L_\nabla \ll 1$ and $l/d \ll 1$. These equations are to first order in these parameters

$$\omega_k \hat{\rho}_k^+(\mathbf{R}_1) = -\frac{\partial}{\partial R_{1\nu}} \hat{p}_{k\nu}^+(\mathbf{R}_1) + \left[\hat{p}_{k\alpha}^+(\mathbf{R}_1) - u_\alpha \hat{\rho}_k^+(\mathbf{R}_1)\right] \frac{\partial}{\partial R_{1\beta}} \eta_B \Delta_{\alpha\beta,\gamma\nu} \frac{\partial u_\nu}{\partial R_{1\gamma}}$$
$$+ \frac{\rho}{nk_B T} \frac{\hat{T}_k^+(\mathbf{R}_1)}{T} \left[\frac{\partial}{\partial R_{1\alpha}} \lambda_B \frac{\partial T}{\partial R_{1\alpha}} + 2\eta_B D_{\alpha\beta}\left(D_{\alpha\beta} - \frac{\delta_{\alpha\beta}}{3} D_{\gamma\gamma}\right)\right] \tag{2.20a}$$

and

$$-\omega_k \hat{p}_{k\alpha}^+(\mathbf{R}_1) = \frac{\partial}{\partial R_{1\alpha}} \left[\frac{k_B T}{m} \hat{\rho}_k^+(\mathbf{R}_1) + nk_B \hat{T}_k^+(\mathbf{R}_1)\right]$$
$$- \frac{\hat{\rho}_k^+(R_1)}{\rho} \frac{\partial(nk_B T)}{\partial R_{1\alpha}}$$
$$+ \frac{k_B T}{m} \frac{\partial}{\partial R_{1\beta}} \eta_B \Delta_{\alpha\beta,\gamma\nu} \frac{\partial}{\partial R_{1\gamma}} \frac{\left[\hat{p}_{k\nu}^+(\mathbf{R}_1) - u_\nu \hat{\rho}_k^+(\mathbf{R}_1)\right]}{nk_B T}$$
$$- \frac{5}{2} nk_B \frac{\partial T}{\partial R_{1\alpha}} \frac{\hat{T}_k^+(\mathbf{R}_1)}{T}$$
$$- 2T \frac{\partial}{\partial R_{1\beta}} \eta_B \Delta_{\alpha\beta,\gamma\nu} \frac{\partial u_\nu}{\partial R_{1\gamma}} \frac{\hat{T}_k^+(\mathbf{R}_1)}{T}$$
$$- \left[\hat{p}_{k\beta}^+(\mathbf{R}_1) - u_\beta \hat{\rho}_k^+(\mathbf{R}_1)\right] \frac{\partial u_\beta}{\partial R_{1\alpha}}$$
$$+ u_\beta \frac{\partial}{\partial R_{1\beta}} \left[\hat{p}_{k\alpha}^+(\mathbf{R}_1) - u_\alpha \hat{\rho}_k^+(\mathbf{R}_1)\right]$$
$$- \frac{\left[\hat{p}_{k\alpha}^+(\mathbf{R}_1) - u_\alpha \hat{\rho}_k^+(\mathbf{R}_1)\right]}{nk_B T} u_\beta \frac{\partial(nk_B T)}{\partial R_{1\beta}} - u_\alpha \omega_k \hat{\rho}_k^+(\mathbf{R}_1) \tag{2.20b}$$

and

$$
\begin{aligned}
&-\omega_k \frac{3}{2} n k_B \hat{T}_k^+(\mathbf{R}_1) \\
&\quad = n k_B T \frac{\partial}{\partial R_{1\nu}} \frac{\left[\hat{p}_{k\nu}^+(\mathbf{R}_1) - u_\nu \hat{\rho}_k^+(\mathbf{R}_1)\right]}{\rho} + \frac{3}{2} n k_B u_\alpha \frac{\partial}{\partial R_{1\alpha}} \hat{T}_k^+(\mathbf{R}_1) \\
&\quad - \frac{k_B}{m}\left[\hat{p}_{k\nu}^+(\mathbf{R}_1) - u_\nu \hat{\rho}_k^+(\mathbf{R}_1)\right] \frac{\partial T}{\partial R_{1\nu}} - 3 n k_B \frac{\hat{T}_k^+(\mathbf{R}_1)}{T} u_\alpha \frac{\partial T}{\partial R_{1\alpha}} \\
&\quad - n k_B \hat{T}_k^+(\mathbf{R}_1) \frac{\partial u_\alpha}{\partial R_{1\alpha}} + T^2 \lambda_B \frac{\partial^2}{\partial R_{1\alpha} \partial R_{1\alpha}} \frac{\hat{T}_k^+(\mathbf{R}_1)}{T^2} \\
&\quad - \frac{k_B T^2}{m} \left(\frac{\partial \eta_B}{\partial T}\right)_\rho \frac{\partial u_\alpha}{\partial R_{1\beta}} \\
&\quad \times \Delta_{\alpha\beta,\gamma\nu} \left[\frac{\partial}{\partial R_{1\gamma}} \frac{\left[\hat{p}_{k\nu}^+(\mathbf{R}_1) - u_\nu \hat{\rho}_k^+(\mathbf{R}_1)\right]}{n k_B T} - \frac{m}{k_B} \frac{\hat{T}_k^+(\mathbf{R}_1)}{T^2} \frac{\partial u_\nu}{\partial R_{1\gamma}} \right]
\end{aligned}
\tag{2.20c}
$$

In Eqs. (2.20) all hydrodynamic quantities are to be evaluated at the point $\mathbf{R}_1$.

Boundary conditions for these equations can be obtained, as they were for the right eigenfunction equations, by using Eq. (2.18c) in place of Eq. (2.6e). For diffusive reflection of the particles at the walls we obtain

$$
\begin{aligned}
\hat{p}_{k\nu}^+(\mathbf{R}_w) &= 0 \\
\hat{T}_{k\nu}^+(\mathbf{R}_w) &= 0
\end{aligned}
\tag{2.21}
$$

Equations (2.19), (2.20), and (2.21) form a closed set of equations by which the adjoint hydrodynamic modes can be determined.

Finally, we remark that for slowly varying phenomena the set of hydrodynamic eigenfunctions approximately satisfies the completeness relation:

$$
1 \approx \sum_j |f_l(1)\Theta_j^R(1))(\Theta_j^L(1)| \tag{2.22}
$$

where j is a general eigenfunction index that can be continuous. In Sections 4 and 6 this relation will be used to calculate the singular part of the correlation functions in a gas near the convective instability point at $R = R_c$.

3. THE HYDRODYNAMIC EIGENMODES FOR $R < R_c$

In this section and in Section 5, the general results of Section 2 are used to calculate explicitly the hydrodynamic modes for a fluid in a Bénard cell. Before we can do so, however, we must first specify more precisely the *average* stationary state of the fluid. The average stationary state is described by $f_1(1)$, which is given by the Eqs. (2.3) as far as its velocity dependence is concerned and by the Navier–Stokes equations as far as its position dependence is concerned through $n(\mathbf{R}_1)$, $\mathbf{u}(\mathbf{R}_1)$, and $T(\mathbf{R}_1)$. Because of the nonlinearity of the Boltzmann equation and the ensuing nonlinearity of the Navier–Stokes equations not all solutions of these equations are stable. To find the stable solutions, we proceed as follows, in close parallel to what is done in hydrodynamics.[5] Linearizing the time-dependent nonlinear Boltzmann equation[16] for $f_1(1t)$ [cf. Eq. (2.1c)] around the steady state Chapman–Enskog solution $f_1^{\text{CH-E}}$, Eqs. (2.3), by writing $f_1(1t) = f_1^{\text{CH-E}}(1) + \delta f_1(1t)$, one is led to a linear equation for $\delta f_1(1t)$ of the form

$$\left[\frac{\partial}{\partial t} + L_{ss}(1) - \bar{T}_w(1)\right]\delta f_1(1t) = 0$$

The stability of the stationarity state, $f_1^{\text{CH-E}}(1)$, then follows if all the eigenvalues, ω_j, of the operator $[L_{ss}(1) - \bar{T}_w(1)]$ are greater than zero since $\delta f_1(1t)$ would then decay to zero as $t \to \infty$. Therefore, since we are only interested in the hydrodynamic regime, i.e., small ω_j, the study of the stability of the $f_1^{\text{CH-E}}(1)$ reduces to the study of Eqs. (2.13), i.e., of the eigenvalue problem obtained by linearizing the Navier–Stokes equations around the stationary state described kinetically by $f_1^{\text{CH-E}}(1)$.

As mentioned above, an explicit expression for $f_1(1)$ can be derived by using Eqs. (2.3) for its velocity dependence and by using the Navier–Stokes equations to determine its position dependence through $n(\mathbf{R}_1)$, $\mathbf{u}(\mathbf{R}_1)$ and $T(\mathbf{R}_1)$. When $R < R_c$, the Navier–Stokes equations that describe the stable average state are according to the Chapman–Enskog method

$$\mathbf{u} = 0 \tag{3.1a}$$

$$\frac{d}{dz_1}(nk_B T) = \rho g \tag{3.1b}$$

$$\frac{d}{dz_1}\left(\lambda_B \frac{dT}{dz_1}\right) = 0 \tag{3.1c}$$

In giving the Eqs. (3.1), we assumed that the gravitational field and the varying temperature field are in the z direction. Further, we take the fluid to be of infinite extent in the x, y plane, but bounded in the z direction by parallel plates at $z_1 = 0$ and d.

To prove that the solutions of the Eqs. (3.1) represent the stable state of the fluid for $R < R_c$, one has to establish that the eigenvalues ω_j in Eq. (2.4) are all greater than zero. That is, we have to consider the hydrodynamic eigenvalue problem given in Section 2, Eqs. (2.13) or (2.20). The equations for the right eigenvalue problem are then [cf. Eq. (2.13)]

$$\omega_j \hat{\rho}_j(\mathbf{R}_1) = \frac{\partial}{\partial R_{1\nu}} \hat{p}_{j\nu}(\mathbf{R}_1) \tag{3.2a}$$

$$-\omega_j \hat{p}_{j\alpha}(\mathbf{R}_1) = -\frac{\partial}{\partial R_{1\alpha}}\left[nk_B \hat{T}_j(\mathbf{R}_1) + \frac{k_B T}{m}\hat{\rho}_j(\mathbf{R}_1)\right] + \delta_{\alpha z} g \hat{\rho}_j(\mathbf{R}_1) + \frac{\partial}{\partial R_{1\beta}} \eta_B \Delta_{\alpha\beta,\gamma\nu} \frac{\partial}{\partial R_{1\gamma}} \frac{\hat{p}_{j\nu}(\mathbf{R}_1)}{\rho} \tag{3.2b}$$

$$-\omega_j \frac{3}{2} nk_B \hat{T}_j(\mathbf{R}_1) = -\frac{3}{2}\frac{k_B}{m}\hat{p}_{jz}(\mathbf{R}_1)\frac{dT}{dz_1} - nk_B T \frac{\partial}{\partial R_{1\nu}} \frac{\hat{p}_{j\nu}(\mathbf{R}_1)}{\rho} + \frac{\partial}{\partial R_{1\alpha}} \lambda_B \frac{\partial}{\partial R_{1\alpha}} \hat{T}_j(\mathbf{R}_1) + \frac{\partial}{\partial z_1}\left(\frac{\partial \lambda_B}{\partial T}\right)_\rho \hat{T}_j(\mathbf{R}_1)\frac{dT}{dz_1} \tag{3.2c}$$

To solve the eigenvalue problem defined by Eqs. (3.2), we first order these equations taking into account that there are two small parameters at our disposal. The first small parameter, l/d, which we have already considered before, represents the relative smallness of the terms in Eq. (3.2) containing transport coefficients η_B, λ_B ($\sim l$) to those that do not. The second small parameter is d/L_∇, which we have not used before. Here L_∇ is a gradient length on the order of $T/|\nabla T|$ with $|\nabla T| = dT/dz_1$. This small parameter represents the variation of $\hat{\rho}$, $\hat{T}$, and $\hat{\mathbf{p}}$ (over d) to the relatively slow variations of the hydrodynamic fields (over L_∇).[9] Using Eq. (3.2) and neglecting terms that are second order in these small parameters and using that the eigenvalues, ω_j, of interest are of order lc/d^2 [10] yields the equations

$$\frac{\partial \hat{p}_{j\nu}(\mathbf{R}_1)}{\partial R_{1\nu}} = 0 + O(l/d) \tag{3.3a}$$

$$\hat{\rho}_j(\mathbf{R}_1) = -\frac{\rho}{T}\hat{T}_j(\mathbf{R}_1) + O(l/d) \tag{3.3b}$$

[9] For typical experiments in a Bénard cell $d = 0.1$ cm, $T = 300$ K and $dT/dz_1 = 50$ K/cm, which yields $d/L_\nabla \sim 1/60 \ll 1$.

[10] In giving this estimate, we are neglecting sound mode phenomena, whose lowest-order eigenvalues are of $O(c/d)$, with c the velocity of sound (cf. Section 7 for a discussion). Further, we used that, away from the instability point, the eigenvalues of interest are at most of $O(lc/d^2)$.

$$-\omega_j\hat{p}_{j\alpha}(\mathbf{R}_1) = -\frac{\partial}{\partial R_{1\alpha}}\left\{nk_B\hat{T}_j(\mathbf{R}_1) + \frac{k_BT}{m}\hat{\rho}_j(\mathbf{R}_1)\right\} - \delta_{\alpha z}\rho g\frac{\hat{T}_j(\mathbf{R}_1)}{T} + \nu\frac{\partial^2}{\partial R_{1\beta}\partial R_{1\beta}}\hat{p}_{j\alpha}(\mathbf{R}_1) \tag{3.3c}$$

$$-\omega_j\hat{T}_j(\mathbf{R}_1) = -\frac{\hat{p}_{jz}(\mathbf{R}_1)}{\rho}\frac{dT}{dz_1} + D_T\frac{\partial^2\hat{T}_j(\mathbf{R}_1)}{\partial R_{1\beta}\partial R_{1\beta}} \tag{3.3d}$$

In giving Eqs. (3.3), we have neglected all sound mode phenomena (cf. Section 7.2). Here the kinematic viscosity $\nu \equiv \eta/\rho$ equals in our case of a dilute gas η_B/ρ, while the thermal diffusivity $D_T \equiv \lambda/\rho C_P$ equals $2\lambda/5nk_B$. In Eqs. (3.3a) and (3.3b), we have explicitly exhibited that the corrections are of $O(l/d)$ since these estimates are used in obtaining Eqs. (3.3c) and (3.3d). Furthermore, the ordering scheme presented here enables us to consider $\rho g/T$, $dT/dz_1\rho$, ν, and D_T in Eqs. (3.3c) and (3.3d) as constants since their spatial variations would lead to terms which are second order in the small parameters l/d and d/L_∇.

The system of equations (3.3) are subject to boundary conditions. Although those given by Eq. (2.15b) are the most realistic, we will use in this paper the mathematically simpler free–free boundary conditions:

$$\hat{T}_j = \hat{p}_{jz} = \partial\hat{p}_{jx}/\partial z_1 = \partial\hat{p}_{jy}/\partial z_1 = 0 \quad \text{at} \quad z_1 = 0 \text{ and } d \tag{3.4}$$

that are often used in the literature on the Bénard problem.[5-11,19] With Eqs. (3.3) and (3.4), the right eigenvalue problem can be solved.[5] There is one viscous eigenvalue:

$$\omega_\nu = \nu\left(k_{\|}^2 + k_z^2\right) \equiv \nu k^2 \tag{3.5a}$$

with expansion coefficients [cf. Eq. (2.11)] given by

$$\hat{\rho}_\nu(\mathbf{R}_1, k_z, \mathbf{k}_{\|}) = \hat{T}_\nu(\mathbf{R}_1, k_z, \mathbf{k}_{\|}) = \hat{p}_{\nu z}(\mathbf{R}_1, k_z, \mathbf{k}_{\|}) = 0$$

$$\hat{p}_{\nu x}(\mathbf{R}_1, k_z, \mathbf{k}_{\|}) = \frac{e^{i\mathbf{k}_{\|}\cdot\mathbf{R}_{1_{\|}}}}{2\pi}A_\nu(k_z, k_{\|})\hat{k}_y\cos k_z z_1 \tag{3.5b}$$

$$\hat{p}_{\nu y}(\mathbf{R}_1, k_z, \mathbf{k}_{\|}) = -\frac{e^{i\mathbf{k}_{\|}\cdot\mathbf{R}_{1_{\|}}}}{2\pi}A_\nu(k_z, k_{\|})\hat{k}_x\cos k_z z_1$$

In Eq. (3.5) ν, k_z, and $\mathbf{k}_{\|}$ are all eigenfunction labels and the wave numbers $\mathbf{k} = (k_x, k_y, k_z)$, $\mathbf{k}_{\|} = (k_x, k_y)$ and $\hat{k} = \mathbf{k}/|\mathbf{k}|$ have been defined with $k_z = n\pi/d$ $(n = 0, 1, 2, \ldots)$ discrete and $\mathbf{k}_{\|}$ continuous. Further, $A_\nu(k_z, k_{\|})$ is a normalizing factor that will be determined later and $\mathbf{R}_{1_{\|}} = (x_1, y_1)$. In addition to this eigenvalue and eigenfunction, there are two others that are combinations of the equilibrium heat and viscous modes. The eigenvalues for these modes are

$$\omega_{\lambda\pm}(k_z,k_{\|}) \equiv \lambda_{\pm}(k_z,k_{\|})$$
$$= \frac{(\nu+D_T)k^2}{2}\left\{1 \pm \left[1 - \frac{4\nu D_T}{(\nu+D_T)^2}\left(1 - \frac{R(k_z,k_{\|})}{R_c}\right)\right]^{1/2}\right\} \tag{3.6a}$$

where

$$\frac{R(k_z,k_{\|})}{R_c} \equiv \frac{g\alpha_T(dT/dz_1)k_{\|}^2}{\nu D_T k^6} \tag{3.6b}$$

The expansion coefficients [cf. Eq. (2.11)] for these modes are

$$\hat{\rho}_{\lambda_\pm}(\mathbf{R}_1,k_z,\mathbf{k}_{\|}) = -\frac{e^{i\mathbf{k}_{\|}\cdot\mathbf{R}_{1_{\|}}}}{2\pi}\frac{d\log T}{dz_1}\frac{A_{\lambda_\pm}(k_z,k_{\|})\sin k_z z_1}{\left[\lambda_{\pm}(k_z,k_{\|}) - D_T k^2\right]}$$

$$\hat{T}_{\lambda_\pm}(\mathbf{R}_1,k_z,\mathbf{k}_{\|}) = \frac{e^{i\mathbf{k}_{\|}\cdot R_{1_{\|}}}}{2\pi}\frac{1}{\rho}\frac{dT}{dz_1}\frac{A_{\lambda_\pm}(k_z,k_{\|})\sin k_z z_1}{\left[\lambda_{\pm}(k_z,k_{\|}) - D_T k^2\right]}$$

$$\hat{p}_{\lambda_\pm z}(\mathbf{R}_1,k_z,\mathbf{k}_{\|}) = \frac{e^{i\mathbf{k}_{\|}\cdot\mathbf{R}_{1_{\|}}}}{2\pi}A_{\lambda_\pm}(k_z,k_{\|})\sin k_z z_1 \tag{3.6c}$$

$$\hat{p}_{\lambda_\pm y}(\mathbf{R}_1,k_z,\mathbf{k}_{\|}) = \frac{e^{i\mathbf{k}_{\|}\cdot\mathbf{R}_{1_{\|}}}}{2\pi}A_{\lambda\pm}(k_z,k_{\|})\frac{ik_y k_z}{k^2}\cos k_z z_1$$

$$\hat{p}_{\lambda_\pm x}(\mathbf{R}_1,k_z,\mathbf{k}_{\|}) = \frac{e^{i\mathbf{k}_{\|}\cdot\mathbf{R}_{1_{\|}}}}{2\pi}A_{\lambda_\pm}(k_z,k_{\|})\frac{ik_x k_z}{k^2}\cos k_z z_1$$

with k_z, $\mathbf{k}_{\|}$, $\mathbf{k}$, and $\mathbf{R}_{1_{\|}}$ given above Eq. (3.6a) and where $A_{\lambda_\pm}(k_z,k_{\|})$ is a normalizing factor that will be determined later. Further, in giving Eq. (3.6), we have used the inequality $mg/k_B T \ll d(\log T)/dz_1$.

We remark that the eigenvalue $\lambda_-(k_z,k_{\|})$ becomes zero when $R(k_z,k_{\|})/R_c = 1$ and it is at this point that an arbitrary perturbation no longer decays to zero and the instability occurs. This zero eigenvalue first appears for $k_z = \pi/d$ and $k_{\|} = k_{\|c} = \pi/d\sqrt{2}$, where $k_{\|c}$ is the critical horizontal wave number.[5] Using these wave numbers, the critical Rayleigh number for our boundary conditions can be determined to be $R_c = 27\pi^4/4$.[5]

From Eqs. (2.11), (3.5), and (3.6) one can immediately obtain the right hydrodynamic eigenmodes to $O(\mu^0)$.[7] There is one viscous eigenfunction:

$$f_l(1)\tilde{\Theta}_\nu^R(1,k_z,\mathbf{k}_{\|}) = \frac{e^{i\mathbf{k}_{\|}\cdot\mathbf{R}_{1_{\|}}}}{2\pi}\beta A_\nu(k_z,k_{\|})\left[\hat{k}_y V_{1x} - \hat{k}_x V_{1y}\right]\cos(k_z z_1)\phi_l(1) \tag{3.7a}$$

with an eigenvalue given by Eq. (3.5a). We remark that this viscous mode will not contribute to the ddcf [cf. (4.9a)] because of tensorial symmetry. Further, the hydrodynamic modes with eigenvalues given by Eq. (3.6a) are

$$f_l(1)\tilde{\Theta}^R_{\lambda_\pm}(1,k_z,\mathbf{k}_\parallel)$$

$$= \frac{e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_{1_\parallel}}}{2\pi m} A_{\lambda_\pm}(k_z,k_\parallel)\phi_l(1)\left\{\frac{d\log T}{dz_1}\frac{\sin(k_z z_1)}{\left[\lambda_\pm(k_z,k_\parallel)-D_T k^2\right]}\right.$$
$$\times\left(\frac{\beta m V_1^2}{2}-\frac{5}{2}\right)+\beta m\sin(k_z z_1)V_{1z}$$
$$\left.+\frac{\beta m\cos(k_z z_1)}{k^2}ik_z\left[k_y V_{1y}+k_x V_{1x}\right]\right\} \tag{3.7b}$$

To determine the normalizing factors A_ν and $A_{\lambda_\pm}$ in Eq. (3.7) and to complete the calculation of the modes for $R < R_c$, the adjoint eigenfunctions, $\Theta_j^L(1)$, are needed. The hydrodynamic equations for the left eigenvalue problem when $R < R_c$ are [cf. Eq. (2.20)]

$$\omega_j\hat{\rho}_j^+(\mathbf{R}_1) = \frac{\partial}{\partial R_{1\alpha}}\hat{p}_{j\alpha}^+(\mathbf{R}_1) \tag{3.8a}$$

$$-\omega_j\hat{p}_{j\alpha}^+(\mathbf{R}_1) = \frac{\partial}{\partial R_{1\alpha}}\left[\frac{k_B T}{m}\hat{\rho}_j^+(\mathbf{R}_1)+nk_B\hat{T}_j^+(\mathbf{R}_1)\right]-\hat{\rho}_j^+(\mathbf{R}_1)g\delta_{\alpha z}$$
$$+\frac{k_B T}{m}\frac{\partial}{\partial R_{1\beta}}\eta_B\Delta_{\alpha\beta,\gamma\nu}\frac{\partial}{\partial R_{1\gamma}}\frac{\hat{p}_{j\nu}^+(\mathbf{R}_1)}{nk_B T}$$
$$-\frac{5}{2}nk_B\frac{dT}{dz_1}\frac{\hat{T}_j^+(\mathbf{R}_1)}{T}\delta_{\alpha z} \tag{3.8b}$$

$$-\frac{3}{2}nk_B\omega_j\hat{T}_j^+(\mathbf{R}_1) = nk_B T\frac{\partial}{\partial R_{1\alpha}}\frac{\hat{p}_{j\alpha}^+(\mathbf{R}_1)}{\rho}-\frac{k_B}{m}\hat{p}_{jz}^+(\mathbf{R}_1)\frac{dT}{dz_1}$$
$$+T^2\lambda_B\frac{\partial^2}{\partial R_{1\alpha}\partial R_{1\alpha}}\frac{\hat{T}_j^+(\mathbf{R}_1)}{T^2} \tag{3.8c}$$

The system of equations given by Eqs. (3.8) can be solved in a manner analogous to that used to determine the right eigenfunctions. With the left eigenfunctions determined and the normalization condition

$$\left(\Theta_k^L(1,k_z,\mathbf{k}_\parallel)f_l(1)\Theta_j^R(1,k_z',\mathbf{k}_\parallel')\right) = \delta_{jk}\delta_{k_z k_z'}\delta(\mathbf{k}_\parallel-\mathbf{k}_\parallel') \tag{3.9}$$

the normalized left and right eigenfunctions can be found.

Since we have neglected the sound eigenfunctions,[10] the relevant modes to leading order in our small parameters are (1) a viscous mode with normalized right and left kinetic eigenfunctions given by

$$f_l(1)\tilde{\Theta}_\nu^R(1,k_z,\mathbf{k}_\parallel) = (2/d)^{1/2}\frac{e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_{1\parallel}}}{2\pi}\left[\beta\rho/\left(\hat{k}_x^2+\hat{k}_y^2\right)\right]^{1/2} \times\left[\hat{k}_y V_{1x} - \hat{k}_x V_{1y}\right]\cos(k_z z_1)\phi_l(1) \tag{3.10a}$$

$$\Theta_\nu^L(1,k_z,\mathbf{k}_\parallel) = (2/d)^{1/2}\frac{e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}_{1\parallel}}}{2\pi}\left[\beta m/n\left(\hat{k}_x^2+\hat{k}_y^2\right)\right]^{1/2} \times\left[\hat{k}_y V_{1x} - \hat{k}_x V_{1y}\right]\cos(k_z z_1) \tag{3.10b}$$

and with an eigenvalue given by Eq. (3.5a); (2) and (3) two eigenfunctions that are combinations of the equilibrium heat and viscous modes. The normalized right and left eigenfunctions for these modes are[11]

$$f_l(1)\tilde{\Theta}_{\lambda_\pm}^R(1,k_z,\mathbf{k}_\parallel) = \frac{e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_{1\parallel}}}{2\pi}\left\{\frac{2}{dm}\,\frac{2}{5}\,\frac{gk_\parallel^2}{k^2}\,\frac{1}{\left[2\lambda_\pm(k_z,k_\parallel)-(\nu+D_T)k^2\right]}\right\}^{1/2} \times\left\{\frac{d\log T}{dz_1}\,\frac{\sin(k_z z_1)}{\left[\lambda_\pm(k_z,k_\parallel)-D_T k^2\right]}\left(\frac{\beta m V_1^2}{2}-\frac{5}{2}\right)+\beta m\sin(k_z z_1)V_{1z} + \beta m\cos(k_z z_1)\frac{ik_z}{k_\parallel^2}\left(k_y V_{1y}+k_x V_{1x}\right)\right\}\phi_l(1) \tag{3.10c}$$

$$\Theta_{\lambda_\pm}^L(1,k_z,\mathbf{k}_\parallel) = \frac{e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}_{1\parallel}}}{2\pi}\left\{\frac{2}{d}\,m\,\frac{2}{5}\,\frac{gk_\parallel^2}{k^2}\,\frac{1}{\left[2\lambda_\pm(k_z,k_\parallel)-(\nu+D_T)k^2\right]}\right\}^{1/2} \times\left\{\sin(k_z z_1)\left(\frac{\beta m V_1^2}{2}-\frac{5}{2}\right)+\frac{5}{2}\,\frac{d\log T}{dz_1}\,\frac{k_\parallel^2}{k^2}\,\frac{\sin(k_z z_1)}{\left[\lambda_\pm(k_z,k_\parallel)-\nu k^2\right]}V_{1z} - \frac{5}{2}\,\frac{d\log T}{dz_1}\,\frac{ik_z}{k^2}\left(k_x V_{1x}+k_y V_{1y}\right)\frac{\cos(k_z z_1)}{\left[\lambda_\pm(k_z,k_\parallel)-\nu k^2\right]}\right\} \tag{3.10d}$$

[11] In giving Eqs. (3.10c) and (3.10d), we have consistently taken ν, D_T, and $d(\log T)/dz_1$ to be constants.

with eigenvalues given by Eqs. (3.6a). Further, an approximate completeness relation exists for these modes given by [cf. Eq. (2.22)]

$$1 \cong \sum_{j=\nu,\lambda_\pm} \sum_{k_z=n\pi/d} \int d\mathbf{k}_\| \big| f_l(1)\Theta_j^R(1,k_z,\mathbf{k}_\|)\big)\big(\Theta_j^L(1,k_z,\mathbf{k}_\|)\big| \tag{3.11}$$

In the next section Eqs. (3.6), (3.9), (3.10), and (3.11) will be used to calculate the singular behavior of the density–density correlation functions in a gas when $R \lesssim R_c$.

4. EQUAL AND UNEQUAL TIME CORRELATION FUNCTIONS FOR $R < R_c$

In this section the results of Section 3 are used to calculate the long-wavelength parts of $G_2(12)$ and $C(1t|2)$ when $R < R_c$. First Eqs. (2.1a) and (2.1d) are solved formally and then the approximate completeness condition given by Eq. (3.11) is used to explicitly evaluate these formal solutions.

The formal solution to Eq. (2.1a) for $C(1t|2)$ when solved as an initial value problem is

$$C(1t|2) = e^{-[L_{ss}(1)-\bar{T}_w(1)]t}\big[\delta(1-2)f_1(1) + G_2(12)\big] \tag{4.1}$$

Here $f_1(1)$ is given to $O(\mu)$ by Eq. (2.3). The formal solution to Eq. (2.1d) for $G_2(12)$ is

$$G_2(12) = \frac{1}{\big[L_{ss}(1) - \bar{T}_w(1) + L_{ss}(2) - \bar{T}_w(2)\big]}\hat{T}(12)(1+P_{12})W(\mathbf{R}_1)W(\mathbf{R}_2)$$
$$\times\big[f_l(2)f_\nabla^{(1)}(1) + O(\mu^2)\big] \tag{4.2}$$

where in giving Eq. (4.2) we have neglected terms of $O(\mu^2)$ on the right-hand side of Eq. (2.1d).[12] Further, for $R < R_c$, $f_\nabla^{(1)}(1)$ as given by Eq. (2.3d), reduces with Eq. (3.1a) to

$$f_\nabla^{(1)}(1) = \frac{1}{\bar{\Lambda}_l(1)} f_l(1)\frac{d\log T}{dz_1}V_{1z}\left(\frac{\beta m V_1^2}{2} - \frac{5}{2}\right) \tag{4.3}$$

Using Eqs. (3.10) and (3.11), Eqs. (4.1) and (4.2) can be explicitly

[12] To the particular solution of Eq. (2.1d) for $G_2(12)$ given by Eq. (4.2), one should, in principle, add the solution of the homogeneous equation $[L_{ss}(1) - \bar{T}_w(1) + L_{ss}(2) - \bar{T}_w(2)]$ $G_2^h(12) = 0$. However, in the low-density approximation considered here, where the difference in position of two colliding particles is neglected, this solution is zero. In general, although $G_2^h(12)$ will not vanish, it will be of very short range, like in thermal equilibrium. It can, therefore, always be neglected when one is interested in the long-range behavior of $G_2(12)$.

evaluated. Inserting one set of modes into Eq. (4.1) yields

$$C(1t\,|\,2) = \sum_j \sum_{k_z} \int d\mathbf{k}_\parallel\, e^{-\omega_j(k_z,k_\parallel)t} \big| f_l(1)\Theta_j^R(1, k_z, \mathbf{k}_\parallel)\big) \times \big(\Theta_j^L(1, k_z, \mathbf{k}_\parallel)\big| \big[\delta(1-2) f_l(1) + G_2(12)\big]\big) \tag{4.4}$$

Using two sets of modes in Eq. (4.2), we obtain

$$G_2(12) = \sum_{j,q} \sum_{k_z, k_z'} \int d\mathbf{k}_\parallel \int d\mathbf{k}_\parallel' \big| f_l(1)\Theta_j^R(1, k_z, \mathbf{k}_\parallel) f_l(2)\Theta_q^R(2, k_z', \mathbf{k}_\parallel')\big)\big) \times \frac{1}{\big[\omega_j(k_z, k_\parallel) + \omega_q(k_z', k_\parallel')\big]} \times \big(\big(\Theta_j^L(1, k_z, \mathbf{k}_\parallel)\Theta_q^L(2, k_z', \mathbf{k}_\parallel')\big| \times \hat{T}(12)(1+P_{12}) W(\mathbf{R}_1) W(\mathbf{R}_2) f_l(2) f_\nabla^{(1)}(1)\big)\big) \tag{4.5}$$

From Eqs. (4.3), (4.5), and the identity[15]

$$\int d\mathbf{V}_1 \int d\mathbf{V}_2\, \Theta_j^L(1, k_z, \mathbf{k}_\parallel)\Theta_q^L(2, k_z', \mathbf{k}_\parallel') \hat{T}(12)(1+P_{12}) W(\mathbf{R}_1) f_l(2) f_\nabla^{(1)}(1) = -\delta(\mathbf{R}_1 - \mathbf{R}_2) W(\mathbf{R}_1) n \frac{d\log T}{dz_1} \int d\mathbf{V}_1\, \phi_l(1)\Theta_j^L(1, k_z, \mathbf{k}_\parallel)\Theta_q^L(1, k_z', \mathbf{k}_\parallel') \times V_{1z}\left(\frac{\beta m V_1^2}{2} - \frac{5}{2}\right) \tag{4.6}$$

the long-wavelength part of $G_2(12)$ can be obtained. Further, this result and Eqs. (2.3a), (4.3), and (4.4) also yield the long-wavelength part of $C(1t\,|\,2)$. Because the final results of these calculations are rather lengthy, we will not reproduce them here. We will, however, explicitly give the unequal time density–density correlation function

$$\langle \delta\rho(\mathbf{R}_1, t)\delta\rho(\mathbf{R}_2)\rangle_{ss} \equiv M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2, t) = m^2 \int d\mathbf{V}_1 \int d\mathbf{V}_2\, C(1t\,|\,2) \tag{4.7}$$

that is observed in light scattering experiments.

Since it is the Fourier transform of the ddcf that is usually measured, we first compute

$$M_{\rho\rho}(k_z, k_z', \mathbf{k}_\parallel, t) = \int d\mathbf{R}_{12} \int_0^d dz_1 \int_0^d dz_2\, e^{-i\mathbf{k}_\parallel \cdot \mathbf{R}_{12_\parallel}} \times \sin(k_z z_1)\sin(k_z' z_2) M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2, t) \tag{4.8a}$$

of which the inverse Fourier transform is

$$M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2, t) = \frac{2}{d} \sum_{k_z, k_z'} \sin(k_z z_1)\sin(k_z' z_2) \int \frac{d\mathbf{k}_\parallel}{(2\pi)^2} e^{i\mathbf{k}_\parallel \cdot \mathbf{R}_{12_\parallel}} \times M_{\rho\rho}(k_z, k_z', \mathbf{k}_\parallel, t) \tag{4.8b}$$

We remark that the Fourier transform $M_{\rho\rho}(k_z, k_z', \mathbf{k}_\|, t)$, defined by Eq. (4.8a), is very convenient to calculate since the eigenfunctions, Eqs. (3.10), are expressed in terms of wave numbers. From Eqs. (2.3a), (3.10), (4.4), (4.5), (4.6), (4.7), and (4.8a) we straightforwardly obtain

$$\begin{aligned} M_{\rho\rho}(k_z, k_z', \mathbf{k}_\|, t) &= \frac{M_{\rho\rho}(k_z, k_z', \mathbf{k}_\|, t=0)}{[\lambda_+(k_z, k_\|) - \lambda_-(k_z, k_\|)]} \\ &\quad \times \{[\nu k^2 - \lambda_-(k_z, k_\|)]\exp[-\lambda_-(k_z, k_\|)t] - [\nu k^2 - \lambda_+(k_z, k_\|)] \\ &\quad \times \exp[-\lambda_+(k_z, k_\|)t]\} + \delta_{k_z k_z'} \frac{\rho k_B T k_\|^2}{(\nu + D_T)k^4} \frac{(\alpha_T dT/dz_1)^2}{[1 - R(k_z, k_\|)/R_c]} \\ &\quad \times \frac{(\exp[-\lambda_-(k_z, k_\|)t] - \exp[-\lambda_+(k_z, k_\|)t])}{[\lambda_+(k_z, k_\|) - \lambda_-(k_z, k_\|)]} \end{aligned} \tag{4.9a}$$

where

$$\begin{aligned} M_{\rho\rho}(k_z, k_z', \mathbf{k}_\|, t=0) &= \delta_{k_z k_z'} \left\{ \frac{\rho^2 k_B T \chi_T(\gamma - 1)}{\gamma} + \frac{\rho k_B T k_\|^2 (\alpha_T dT/dz_1)^2}{D_T(\nu + D_T)k^6[1 - R(k_z, k_\|)/R_c]} \right\} \end{aligned} \tag{4.9b}$$

Here $\gamma = c_p/c_v$ is the ratio of specific heats and $\chi_T = (\partial\rho/\partial p)_T/\rho$ is the isothermal compressibility. We remark that the $M_{\rho\rho}(k_z, k_z', \mathbf{k}_\|, t=0)$ represent Fourier components of the equal time correlation function $\langle \delta\rho(\mathbf{R}_1) \delta\rho(\mathbf{R}_2)\rangle_{ss} \equiv M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2)$.

As mentioned in Section 1, we have written Eqs. (4.9) in a form valid for *all* densities, as obtained from a hydrodynamic treatment rather from the kinetic theory presented here. Of course, the kinetic result is obtained if the low-density limit of Eq. (4.9) is taken, i.e., χ_T is replaced by $(nk_B T)^{-1}$, γ by 5/3, c_p by $\frac{5}{2}kB/m$, α_T by T^{-1}, and the transport coefficients η and λ by their low-density (Boltzmann) values η_B and λ_B as given by Eqs. (2.12a) and (2.12b), respectively.

Examining Eqs. (4.9), we note that when $k_z = \pi/d$ ($n = 1$) and $k_\|$ is near $k_{\|_c} = \pi/d\sqrt{2}$, the equal and unequal time (ddcfs) are singular if $R \cong R_c$. Using

$$1 - \frac{R(k_z = \pi/d, k_\| \cong k_{\|_c})}{R_c} \cong \left(1 - \frac{R}{R_c}\right) + \frac{4(k_\| - k_{\|_c})^2}{3k_{\|_c}^2} \equiv E_<(k_\|) \tag{4.10a}$$

and

$$\lambda_-(k_z = \pi/d,\, k_\| \cong k_{\|_c}) \cong k_c^2 \frac{\nu D_T}{(\nu + D_T)} E_<(k_\|) \tag{4.10b}$$

with $R_c = 27\pi^4/4$ and $k_c^2 = \pi^2/d^2 + k_{\|_c}^2 = 3\pi^2/2d^2$, yields

$$\begin{aligned}
&M_{\rho\rho}(k_z = k_z' = \pi/d,\, k_\| \cong k_{\|_c},\, t) \\
&\quad = \frac{M_{\rho\rho}(k_z = k_z' = \pi/d,\, k_\| \cong k_{\|_c},\, t = 0)}{(\nu + D_T)k_c^2} \\
&\quad \times \left\{ \nu k_c^2 \exp\left[\frac{-\nu D_T k_c^2}{(\nu + D_T)} E_<(k_\|) t \right] + D_T k^2 \exp\left[-(\nu + D_T) k_c^2 t \right] \right\} \\
&\quad + \frac{\rho k_B T k_{\|_c}^2}{(\nu + D_T)^2 k_c^6} \frac{(\alpha_T dT/dz_1)^2}{E_<(k_\|)} \left\{ \exp\left[-\frac{\nu D_T k_c^2}{(\nu + D_T)} E_<(k_\|) t \right] \right. \\
&\quad \left. - \exp\left[-(\nu + D_T) k_c^2 t \right] \right\}
\end{aligned} \tag{4.11a}$$

where

$$\begin{aligned}
&M_{\rho\rho}(k_z = k_z' = \pi/d,\, k_\| \cong k_{\|_c},\, t = 0) \\
&\quad = \frac{\rho^2 k_B T \chi_T(\gamma - 1)}{\gamma} + \frac{\rho k_B T k_{\|_c}^2}{D_T(\nu + D_T) k_c^6} \frac{(\alpha_T dT/dz_1)^2}{E_<(k_\|)}
\end{aligned} \tag{4.11b}$$

From Eqs. (4.10) and (4.11a) it follows that for the wave numbers $k_z = \pi/d$ and $k_\| \cong k_{\|_c} = \pi/d\sqrt{2}$ those parts of the unequal time density–density correlation function proportional to $\exp[-\lambda_- t]$ exhibit critical slowing down as $|R_c - R| \to 0$. Further, the equal time density–density correlation function, given by Eq. (4.11b), is singular for these wave numbers as $|R_c - R| \to 0$. This singular behavior of $M_{\rho\rho}$ comes from the combination $j = q = \omega_{\lambda_-}$ in Eq. (4.5) for $G_2(12)$, leading to a factor $[\omega_{\lambda_-}(k_z, k_\|) + \omega_{\lambda_-}(k_z', k_\|')]^{-1}$ that ultimately reduces to the factor $[1 - R(k_c, k_\|)/R_c]^{-1}$ in Eqs. (4.9), which gives in turn the singular behavior for $k_\| = k_{\|_c}$ at $R = R_c$. Thus, the singularities in G_2 and $M_{\rho\rho}$ at $R = R_c$ are a direct consequence of a mode-coupling contribution to G_2 from two modes that have vanishing eigenvalues at $R = R_c$, i.e., the same modes that cause the convective instability. We remark that the singularity in $M_{\rho\rho}(t = 0)$ is due to the singularity in the nonequilibrium part of $G_2(12)$. This part of $G_2(12)$ has been discussed previously away from the instability point, where it was shown that it was responsible for long-range correlations in a nonequilibrium fluid.[1-4] We note that the *same* expression for $G_2(12)$, Eq. (4.2), that leads to Eq. (4.11b) can also be used, if the effects of gravity and the walls are neglected, to derive both the long time tails of the time correlation functions that determine the transport coefficients[3,12] and the nonexistence of a virial expansion of the transport coefficients.[3,20] Here we see that Eq. (4.2) for $G_2(12)$ also leads to singularities in the equal time correlation functions near a hydrodynamic instability.

We also note that for $g = 0$ and consequently $R(\mathbf{k})/R_c = 0$, Eqs. (4.9a) and (4.9b) for unequal and equal time correlation functions reduce to the expressions for $M_{\rho\rho}(t)$ and $M_{\rho\rho}(t = 0)$ obtained previously [see Ref. 3, Eq. (3.4)], when gravity and walls were neglected. This implies that the unequal and equal time ddcfs and therefore also the line shape and intensity of the central or Rayleigh line of the light scattered by a nonequilibrium fluid are *independent* of the presence of walls.

In the last part of this section, the spatial decay of the equal time ddcf will be examined. We will also show that the equal time density–density correlation function exhibits modified Ornstein–Zernike behavior and becomes singular when R approaches R_c. From Eqs. (4.8b) and (4.9b), we obtain

$$M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2) = \frac{2}{d} \sum_{k_z} \sin(k_z z_1)\sin(k_z z_2) \int \frac{d\mathbf{k}_\parallel}{(2\pi)^2} e^{i\mathbf{k}_\parallel \cdot \mathbf{R}_{12\parallel}}$$

$$\times \left\{ \frac{\rho^2 k_B T \chi_T(\gamma - 1)}{\gamma} + \frac{\rho k_B T k_\parallel^2}{D_T(\nu + D_T)k^6} \right.$$

$$\left. \times \left(\alpha_T \frac{dT}{dz_1}\right)^2 \frac{1}{[1 - R(k_z, k_\parallel)/R_c]} \right\}$$

$$= \frac{\rho^2 k_B T \gamma_T(\gamma - 1)}{\gamma} \delta(\mathbf{R}_1 - \mathbf{R}_2) + D_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2) \qquad (4.12)$$

Because we are interested only in long-range correlations near $R \cong R_c$, the contribution to $M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2)$ proportional to $\delta(\mathbf{R}_1 - \mathbf{R}_2)$ can be neglected. Further, although all the k_z components of $D_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2)$ are of long range as has been discussed previously,[1-4] here we are interested only in those parts of $D_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2)$ that are of longest range ($\gg d$) and are enhanced near $R \cong R_c$. These enhanced contributions to $D_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2)$ occur when $k_z = \pi/d$ ($n = 1$). Denoting this contribution to $D_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2)$, which becomes singular at $R = R_c$, as $D^s_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2)$ and using Eqs. (4.10) and (4.12) yields

$$D^s_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2) = \rho k_B T \frac{1}{D_T(\nu + D_T)} \frac{2}{d} \sin(\pi z_1/d) \sin(\pi z_2/d)$$

$$\times \frac{1}{2\pi} \int_0^\infty dk_\| \frac{k_\|^3}{\left(k_\|^2 + \pi^2/d^2\right)^3}$$

$$\times \frac{1}{\left[(1 - R/R_c) + (4/3)(k_\| - k_{\|_c})^2/k_{\|_c}^2\right]}$$

$$\times \frac{1}{2\pi} \int_0^{2\pi} d\theta\, e^{ik_\| R_{12_\|} \cos\theta} \tag{4.13}$$

The use of Eq. (4.10) in Eq. (4.12) is justified as long as R is sufficiently close to R_c, because then the major contribution to the wave number integral over $k_\|$ comes from values of $k_\|$ restricted to a small region near $k_{\|_c}$. The asymptotic analysis of Eq. (4.12) for large $R_{12_\|}$ is straightforward with the result

$$D^s_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2, R_{12_\|} \gg d)$$

$$\cong \rho k_B T (\alpha_T dT/dz_1)^2 \frac{1}{D_T(D_T + \nu)} \frac{2}{d} \sin(\pi z_1/d) \sin(\pi z_2/d)$$

$$\times \frac{\sqrt{3}}{54} \frac{d^2}{\pi^2} \frac{1}{\left[1 - R/R_c\right]^{1/2}} \left(2/\pi k_{\|_c} R_{12_\|}\right)^{1/2}$$

$$\times \cos\left[k_{\|_c} R_{12_\|} - \pi/4\right] \exp\left[-R_{12_\|}(1 - R/R_c)^{1/2} \frac{\pi}{2d} \sqrt{\frac{3}{2}}\right] \tag{4.14}$$

A discussion of this and other results presented in this section can be found in Section 7. Further, restrictions on the applicability of the theory presented here due to singular mode-coupling contributions to the eigenvalue $\lambda_-(k_z, k_\|)$ are given in Appendix C.

5. THE HYDRODYNAMIC EIGENMODES FOR $R \gtrsim R_c$

For $R > R_c$ the steady nonconvecting state is no longer stable because the eigenvalue $\lambda_-(k_z = \pi/d,\ k_\| \cong k_{\|_c})$, discussed in the previous sections, is less than zero. The physically realized fluid state is typically still a steady state but with macroscopic motion in the form of two-dimensional rolls.[5,19] The precise form of this state is very hard to determine because of the difficulty in solving the nonlinear Navier–Stokes equations. However, if one is interested, like we are, in the behavior near the instability point, then a perturbation expansion in powers of the deviation of R from R_c can be used and the fluid state can be approximately calculated for small values of $R/R_c - 1$. Denoting the deviations from the nonconvecting hydrodynamic fields by $\Delta\rho$, ΔT, u_z, u_x, and u_y and assuming that the axis of the rolls is in the y direction, an approximate calculation, that can be found in the literature,(19) gives these hydrodynamic fields to $O[R/R_c - 1]$ as

$$\Delta\rho(x_1, z_1) = -\frac{\rho}{T}\Delta T(x_1, z_1) \tag{5.1a}$$

$$\Delta T(x_1, z_1) = -9\sqrt{3}\,\frac{d\pi^3}{R}\,\frac{dT_{nc}}{dz_1}(R/R_c - 1)^{1/2}\sin(\pi z_1/d)\cos ax_1 + \frac{d}{\pi}\,\frac{dT_{nc}}{dz_1}\,\frac{(R - R_c)}{R_c}\sin(2\pi z_1/d) \tag{5.1b}$$

$$u_z(x_1, z_1) = \frac{D_T}{d}2\pi\sqrt{3}\,(R/R_c - 1)^{1/2}\sin(\pi z_1/d)\cos ax_1 \tag{5.1c}$$

$$u_x(x_1, z_1) = -\frac{D_T}{d}4\pi(3/2)^{1/2}(R/R_c - 1)^{1/2}\cos(\pi z_1/d)\sin ax_1 \tag{5.1d}$$

$$u_y(x_1, z_1) = 0 \tag{5.1e}$$

where $a = k_{\|_c} = \pi/d\sqrt{2}$ and dT_{nc}/dz_1 denotes the derivative of the temperature field in the absence of convection. Furthermore, R, dT_{nc}/dz_1, and D_T have been taken to be constants in the derivation of Eq. (5.1) which is consistent with the ordering scheme developed below. In determining these hydrodynamic fields free–free boundary conditions have been used:

$$\Delta T = u_z = \partial u_x/\partial z_1 = 0 \qquad \text{for} \quad z_1 = 0 \text{ and } d \tag{5.2}$$

In this section the formal results of Section 2 are used to calculate explicitly the hydrodynamic modes for a Bénard cell when $R \gtrsim R_c$ and the average hydrodynamic state of the fluid is given by Eqs. (5.1).

To determine the hydrodynamic modes, we must solve the hydrodynamic eigenvalue problem, defined in Section 2, for the average hydrodynamic state discussed above. The relevant hydrodynamic equations for the right eigenvalue problem are given by Eqs. (2.13) with $n = n_{nc} + \Delta\rho/m$, $T = T_{nc} + \Delta T$, and $\mathbf{u}$ given by Eqs. (5.1), where n_{nc} and T_{nc} are the number density and temperature in the absence of convection, respectively. To solve this eigenvalue problem, we order Eq. (2.13), taking into account that there are three small parameters for $R \gtrsim R_c$. This ordering scheme is similar to that used in Section 3 to determine the modes for $R < R_c$ where there were two small parameters. The three small parameters for $R \gtrsim R_c$ are (1) l/d; (2) d/L_∇ with $L_\nabla \sim T/|\nabla T_{nc}|$; and (3) Δ which represents the deviations of the hydrodynamic fields from the nonconvecting state [cf. Eqs. (5.1)]. Neglecting terms that are of second order in these small parameters[13] and using that the eigenvalues ω_j of interest are smaller than terms of order lc/d^2, we obtain from Eqs. (2.13) the equations

$$\frac{\partial}{\partial R_{1\alpha}} \hat{p}_{j\alpha}(\mathbf{R}_1) = 0 + O(l/d) \tag{5.3a}$$

$$\hat{\rho}_j(\mathbf{R}_1) = -\frac{\rho}{T} \hat{T}_j(\mathbf{R}_1) + O(l/d) \tag{5.3b}$$

$$\begin{aligned} -\omega_j \hat{p}_{j\alpha}(\mathbf{R}_1) = &-\frac{\partial}{\partial R_{1\alpha}} \left[nk_B \hat{T}_j(\mathbf{R}_1) + \frac{k_B T}{m} \hat{\rho}_j(\mathbf{R}_1) \right] \\ &- u_\beta \frac{\partial}{\partial R_{1\beta}} \hat{p}_{j\alpha}(\mathbf{R}_1) - \hat{p}_{j\beta}(\mathbf{R}_1) \frac{\partial}{\partial R_{1\beta}} u_\alpha \\ &- g \frac{\rho}{T} \hat{T}_j(\mathbf{R}_1) \delta_{\alpha z} + \nu \frac{\partial^2}{\partial R_{1\beta} \partial R_{1\beta}} \hat{p}_{j\alpha}(\mathbf{R}_1) \end{aligned} \tag{5.3c}$$

$$\begin{aligned} -\omega_j \hat{T}_j(\mathbf{R}_1) = &-u_\alpha \frac{\partial}{\partial R_{1\alpha}} \hat{T}_j(\mathbf{R}_1) - \frac{\hat{p}_{jz}}{\rho} \frac{dT_{nc}}{dz_1} - \frac{\hat{p}_{j\alpha}(\mathbf{R}_1)}{\rho} \frac{\partial \Delta T}{\partial R_{1\alpha}} \\ &+ D_T \frac{\partial^2}{\partial R_{1\beta} \partial R_{1\beta}} \hat{T}_j(\mathbf{R}_1) \end{aligned} \tag{5.3d}$$

The boundary conditions that will be used to solve Eqs. (5.3) are

$$\hat{T}_j = \hat{p}_{jz} = \partial \hat{p}_{jx}/\partial z_1 = \partial \hat{p}_{jy}/\partial z_1 = 0 \qquad \text{at} \quad z_1 = 0 \text{ and } d \tag{5.4}$$

We remark that the ordering scheme presented here enables us to consider $\rho g/T$, $(1/\rho) dT_{nc}/dz_1$, ν, and D_T in Eqs. (5.3c) and (5.3d) as constants since their spatial variations would lead to terms of second order in the small parameters.

To construct the solutions to the system of equations given by Eqs. (5.3) and (5.4), it is convenient to put them into dimensionless form. We do this by introducing scaled variables, denoted by primes:[(5,19)]

$$x_\alpha = dx'_\alpha \quad (x_\alpha = x_1, y_1, z_1), \quad u_\alpha = \frac{D_T}{d} u'_\alpha, \quad \delta T = \frac{\nu D_T T}{g d^3} \delta T', \quad \omega_j = \frac{D_T}{d^2} \omega'_j \tag{5.5}$$

$$\hat{\Gamma}'_j = \frac{d^2}{\rho D_T^2} \left[nk_B \hat{T}_j(\mathbf{R}_1) + \frac{k_B T}{m} \hat{\rho}_j(\mathbf{R}_1) \right], \quad \hat{p}_{j\alpha} = \frac{\rho D_T}{d} \hat{p}'_{j\alpha}, \quad \hat{T}_j = \frac{\nu D_T T}{g d^3} \hat{T}'_j$$

Here $P = \nu/D_T$ is the Prandtl number. Using then Eqs. (5.5) and (5.1) in

[13] In this connection it should be noted that the parameter Δ itself has an expansion in powers of $(R - R_c)^{1/2}$ [cf. Eq. (5.1)]. Since we will eventually calculate eigenvalue corrections of $O(R - R_c)$, one might think that terms of $O(\Delta^2)$ should be consistently retained. However, if this is done, then it can be shown that the additional terms lead to contributions of $O[(R - R_c)d/L_\nabla]$, which are small compared to the terms of $O[(R - R_c)]$ that have been kept.

Eqs. (5.3), the scaled equations are, dropping the primes,

$$\frac{\partial}{\partial R_{1\alpha}} \hat{p}_{j\alpha}(\mathbf{R}_1) = 0 + O(l/d) \tag{5.6a}$$

$$\begin{aligned} -\omega_j \hat{p}_{j\alpha}(\mathbf{R}_1) = & -\frac{\partial}{\partial R_{1\alpha}} \hat{\Gamma}_j(\mathbf{R}_1) - \hat{p}_{j\beta}(\mathbf{R}_1) \frac{\partial}{\partial R_{1\beta}} u_\alpha - u_\beta \frac{\partial}{\partial R_{1\beta}} \hat{p}_{j\alpha}(\mathbf{R}_1) \\ & + P \frac{\partial^2}{\partial R_{1\beta} \partial R_{1\beta}} \hat{p}_{j\alpha}(\mathbf{R}_1) - P\delta_{\alpha z} \hat{T}_j(\mathbf{R}_1) \end{aligned} \tag{5.6b}$$

$$\begin{aligned} -\omega_j \hat{T}_j(\mathbf{R}_1) = & -u_\alpha \frac{\partial}{\partial R_{1\alpha}} \hat{T}_j(\mathbf{R}_1) - \hat{p}_{j\alpha}(\mathbf{R}_1) \frac{\partial \delta T}{\partial R_{1\alpha}} - R\hat{p}_{jz}(\mathbf{R}_1) \\ & + \frac{\partial^2}{\partial R_{1\alpha} \partial R_{1\alpha}} \hat{T}_j(\mathbf{R}_1) \end{aligned} \tag{5.6c}$$

with

$$\Delta T(x_1, z_1) = -\epsilon \frac{\sqrt{2}}{2} (\pi^2 + a^2)^2 \sin \pi z_1 \sum_{\sigma = \pm 1} e^{ia\sigma x_1} + \epsilon^2 \frac{(\pi^2 + a^2)^2}{4} a^2 \sin 2\pi z_1 \tag{5.7a}$$

$$u_z(x_1, z_1) = \epsilon \frac{\sqrt{2}}{2} a^2 \sin \pi z_1 \sum_{\sigma = \pm 1} e^{ia\sigma x_1} \tag{5.7b}$$

$$u_x(x_1, z_1) = i\epsilon \frac{\sqrt{2}}{2} \pi a \cos \pi z_1 \sum_{\sigma = \pm 1} \sigma e^{ia\sigma x_1} \tag{5.7c}$$

$$u_y(x_1, z_1) = 0 \tag{5.7d}$$

where $\epsilon \equiv (24/\pi)^{1/2}(R/R_c - 1)^{1/2}$. The boundary conditions on Eqs. (5.6) are, from Eq. (5.4)

$$\hat{T}_j = \hat{p}_{jz} = \frac{\partial \hat{p}_{jx}}{\partial z_1} = \frac{\partial \hat{p}_{jy}}{\partial z_1} = 0 \qquad \text{at} \quad z_1 = 0 \text{ and } 1 \tag{5.8}$$

Inserting Eqs. (5.7) into Eqs. (5.6), we obtain a system of linear differential equations with coefficients that are independent of y_1 and periodic in the x_1 coordinate with a period of $2\pi/a$. Using this and Eq. (5.8), we are motivated to look for right eigenfunctions of the form[21]

$$\hat{T}_j(\mathbf{R}_1) = \frac{1}{2\pi} \sum_{n=0}^{\infty} \sum_{m=-\infty}^{+\infty} e^{i\mathbf{k}_\parallel \cdot \mathbf{R}_{1\parallel} + i\, max_1} \sin(n\pi z_1) \hat{T}_j(n, m) \tag{5.9a}$$

$$\hat{p}_{jz}(\mathbf{R}_1) = \frac{1}{2\pi} \sum_{n=0}^{\infty} \sum_{m=-\infty}^{+\infty} e^{i\mathbf{k}_\parallel \cdot \mathbf{R}_{1\parallel} + i\, max_1} \sin(n\pi z_1) \hat{p}_{jz}(n, m) \tag{5.9b}$$

$$\hat{p}_{j\alpha=y,x}(\mathbf{R}_1) = \frac{1}{2\pi} \sum_{n=0}^{\infty} \sum_{m=-\infty}^{+\infty} e^{ik_\parallel \cdot \mathbf{R}_{1\parallel} + i\, max_1} \cos(n\pi z_1) \hat{p}_{j\alpha=y,x}(n, m) \tag{5.9c}$$

To determine the approximate eigenfunctions and eigenvalues of Eqs. (5.6), one must first insert the Eqs. (5.9) into Eqs. (5.6), eliminating the "pressure" term $\partial\hat{\Gamma}_j(\mathbf{R}_1)/\partial\mathbf{R}_{1\alpha}$ by taking the curl of Eq. (5.6b). One then obtains an infinite set of coupled algebraic equations for the coefficients $\hat{T}_j(n,m)$ and $\hat{p}_{j\alpha}(n,m)$. To decouple and solve these algebraic equations, we assume that the eigenfunctions and the eigenvalues can be expanded in powers of $\epsilon \sim (R/R_c - 1)^{1/2}$. With this solution method one can then compute the eigenfunctions to $O(\epsilon)$ and the eigenvalues to $O(\epsilon^2)$. This method of solution can be found in the literature, where it is used for the determination of the critical eigenmode.[19,21] Here it will be employed to determine other eigenmodes as well. Since the procedure is rather lengthy, we will outline the main steps in Appendix A and give only the results needed for the following sections in the main text.

Apart from a viscous mode, that is identical to that for $R < R_c$ [Eqs. (3.5a), (3.10a), (3.10b)], there are two eigenmodes with eigenvalues $\omega_{\Lambda_\pm} \equiv \Lambda_\pm(N, k_\|)$ $(N = 1, 2, \ldots)$ that are the extensions to $R > R_c$ of those with eigenvalues $\lambda_\pm$ for $R < R_c$. For Λ_+ for all N and for Λ_- for $N \geqslant 2$, one only needs the eigenvalues to $O(\epsilon^0)$. They are

$$\Lambda_\pm^{(0)}(N, k_\|) = (\nu + D_T)\frac{k^2}{2}\left\{1 \pm \left[1 - \frac{4\nu D_T}{(\nu + D_T)^2}\left(1 - \frac{R_c k_\|^2}{d^4 k^6}\right)\right]^{1/2}\right\} \tag{5.10a}$$

where $k^2 = k_\|^2 + N^2\pi^2/d^2$. For Λ_- and $N = 1$, the critical eigenvalue $\Lambda_-(1, \mathbf{k}_\|)$ for $k_\| \cong k_{\|_c} = \pi/d\sqrt{2}$ is needed to $O(\epsilon^2) \sim (R/R_c - 1)$ in order to ensure a well-behaved description for the correlation functions for $\epsilon \sim (R/R_c - 1)^{1/2}$ small but nonzero. One has

$$\Lambda_-\left(1, k_\| \cong k_{\|_c}, \cos\theta\right) \cong \frac{\nu D_T k_c^2}{\nu + D_T}\left[\frac{4}{3}\frac{\left(k_\| - k_{\|_c}\right)^2}{k_{\|_c}^2} + (R/R_c - 1) f(\cos\theta)\right] \tag{5.10b}$$

with $k_c^2 = 3\pi^2/2d^2$ and

$$\begin{aligned} f(\cos\theta) = \frac{1}{P}\sum_{\sigma = \pm 1} & \frac{(1 + \sigma\cos\theta)^2}{\left[(5 - \sigma\cos\theta)^3 - \frac{27}{4}(1 - \sigma\cos\theta)\right]} \\ & \times\left[\frac{27}{4}(1 - \sigma\cos\theta) + P(5 - \sigma\cos\theta)^2\right. \\ & \left. + \frac{3}{P}(1 - \sigma\cos\theta)(5 - \sigma\cos\theta)\right] \end{aligned} \tag{5.10c}$$

Here θ is the angle between k and the direction across the rolls, the $\hat{x}$ axis.

The corresponding hydrodynamic eigenfunctions to lowest order in ϵ, i.e., to $O(\epsilon^0)$, are given by

$$f_l(1)\tilde{\Theta}^{R(0)}_{\Lambda_\pm}(1,N,\mathbf{k}_\|)$$

$$= \frac{e^{i\mathbf{k}_\|\cdot\mathbf{R}_{1\|}}}{2\pi}\left[\frac{2}{dm}\,\frac{2}{5}\,\frac{gk_\|^2}{k^2}\,\frac{1}{\left[2\Lambda^{(0)}_\pm(N,k_\|)-(\nu+D_T)k^2\right]}\right]^{1/2}$$

$$\times\left\{\frac{d\log T_{nc}}{dz_1}\,\frac{\sin(N\pi z_1/d)}{\left[\Lambda^{(0)}_\pm(N,k_\|)-D_Tk^2\right]}\left(\frac{\beta mC_1^2}{2}-\frac{5}{2}\right)\right.$$

$$+\beta mC_{1z}\sin(N\pi z_1/d)$$

$$\left.+\beta m\cos(N\pi z_1/d)\,\frac{iN\pi}{dk_\|^2}\,(k_yC_{1y}+k_xC_{1x})\right\}\phi_l(1) \qquad (5.11\text{a})$$

and

$$\Theta^{L(0)}_{\Lambda_\pm}(1,N,\mathbf{k}_\|) = \frac{e^{-i\mathbf{k}_\|\cdot\mathbf{R}_{1\|}}}{2\pi}\left[\frac{2}{d}\,m\,\frac{2}{5}\,\frac{gk_\|^2}{k^2}\,\frac{1}{\left[2\Lambda^{(0)}_\pm(N,k_\|)-(\nu+D_T)k^2\right]}\right]^{1/2}$$

$$\times\left\{\sin(N\pi z_1/d)\left(\frac{\beta mC_1^2}{2}-\frac{5}{2}\right)\right.$$

$$+\frac{5}{2}\,\frac{d\log T_{nc}}{dz_1}\,\frac{k_\|^2}{k^2}\,\frac{\sin(N\pi z_1/d)}{\left[\Lambda^{(0)}_\pm(N,k_\|)-\nu k^2\right]}\,C_{1z}$$

$$-\frac{5}{2}\,\frac{d\log T_{nc}}{dz_1}\,\frac{iN\pi}{dk^2}\,(k_xC_{1x}+k_yC_{1y})$$

$$\left.\times\frac{\cos(N\pi z_1/d)}{\left[\Lambda^{(0)}_\pm(N,k_\|)-\nu k^2\right]}\right\} \qquad (5.11\text{b})$$

We remark that to lowest order in ϵ the eigenfunctions for $R \gtrsim R_c$ have the same form as the eigenfunctions given by Eq. (3.10) for $R < R_c$. As discussed in Section 7(5), this is not anymore true in the next order in ϵ. Also, the nonvanishing eigenvalues have the same form for $R > R_c$ and $R < R_c$ and it is only the eigenvalue $\Lambda_-(1,\mathbf{k}_\|)$ that must be modified to take into account the presence of the two-dimensional convection.

The completeness relation for these modes is

$$1 \cong \sum_{j=\Lambda_{\pm},\nu} \sum_{N=1,\ldots} \int d\mathbf{k}_{\parallel} \, |f_l(1)\Theta_j^R(1,N,\mathbf{k}_{\parallel}))(\Theta_j^L(1,N,\mathbf{k}_{\parallel})| \qquad (5.12)$$

In the next section Eqs. (5.10), (5.11), and (5.12) will be used to calculate the singular correlation functions that exist in a gas when $R \gtrsim R_c$.

6. EQUAL AND UNEQUAL TIME CORRELATION FUNCTIONS FOR $R \gtrsim R_c$

In this section the results of Section 5 will be used to calculate the long-wavelength parts of $G_2(12)$ and $C(1t|2)$ to lowest order in $\epsilon \sim (R/R_c - 1)^{1/2}$ when $R \gtrsim R_c$. We will follow the procedure given in Section 4, i.e., first Eqs. (2.1a) and (2.1d) are solved formally and then the approximate complete set given by Eq. (5.12) will be used to explicitly evaluate these formal solutions. Because the method is identical to that given in Section 4, we will be brief.

We first remark that to lowest order in ϵ we can neglect the term in Eq. (2.3d) proportional to $\partial u_\alpha/\partial R_{1\beta}$, since $u \sim O(\epsilon)$, and write the correction to the local Maxwellian distribution function as

$$f_\nabla^{(1)}(1) = \frac{1}{\overline{\Lambda}_l(1)} f_l(1) \frac{d \log T_{nc}}{dz_1} V_{1z}\left(\frac{\beta m V_1^2}{2} - \frac{5}{2}\right) \qquad (6.1)$$

Using Eq. (6.1), we now evaluate $G_2(12)$ and $C(1t|2)$ to lowest order in ϵ.

The formal solutions to Eqs. (2.1a) and (2.1d) for $C(1t|2)$ and $G_2(12)$ are still given by Eqs. (4.1) and (4.2), respectively. Inserting one set of modes, given by Eqs. (5.11) for $R \gtrsim R_c$, into Eq. (4.1) and neglecting the viscous mode, which does not contribute to the ddcfs, yields

$$\begin{aligned} C^{(0)}(1t|2) = & \sum_{\sigma=\pm 1} \sum_{N=1,\ldots} \int d\mathbf{k}_{\parallel} \\ & \times \exp\left\{-\left[\Lambda_\sigma^{(0)}(N,k_{\parallel}) + \delta_{\sigma,-1}\delta_{N1}\Lambda_\sigma^{(2)}(N,\mathbf{k}_{\parallel})\right]t\right\} \\ & \times \left|f_l(1)\Theta_{\Lambda_\sigma}^{R^{(0)}}(1,N,\mathbf{k}_{\parallel})\right) \\ & \times \left(\Theta_{\Lambda_\sigma}^{L^{(0)}}(1,N,\mathbf{k}_{\parallel})\right| \left[\delta(1-2)f_1^{(0)}(1) + G_2^{(0)}(12)\right]\Big) \end{aligned} \qquad (6.2)$$

where the superscript (0) denotes that the lowest-order approximation in ϵ

is taken. Using two sets of modes, given by Eqs. (5.11), in Eq. (4.2) yields

$$G_2^{(0)}(12) = \sum_{j,i=\Lambda_\pm} \sum_{N,M} \int d\mathbf{k}_\parallel \int d\mathbf{k}'_\parallel \big| f_l(1)\Theta_j^{R^{(0)}}(1,N,\mathbf{k}_\parallel) f_l(2)\Theta_i^{R^{(0)}}(2,M,\mathbf{k}_\parallel)\big)\big)$$
$$\times \left[\omega_j(N,k_\parallel) + \omega_i(M,k'_\parallel)\right]^{-1}$$
$$\times \big(\big(\Theta_j^{L^{(0)}}(1,N,\mathbf{k}_\parallel)\Theta_i^{L^{(0)}}(2,M,\mathbf{k}'_\parallel)\,\big|$$
$$\times \hat{T}(12)(1+P_{12})W(\mathbf{R}_1)W(\mathbf{R}_2)f_l(2)f_\nabla^{(1)}(1)\big)\big) \tag{6.3}$$

From Eqs. (6.1) and (6.3) and a relation almost identical to that given by Eq. (4.6), the long wavelength part of $G_2^{(0)}(12)$ can be obtained. Using this and Eqs. (2.3a), (6.1), and (6.2), the long wavelength part of $C^{(0)}(1t\,|\,2)$ then follows. Again, we will not reproduce this result here, but give only the singular contributions to the ddcf given by Eq. (4.7).

Defining the Fourier transform of the ddcf, $M_{\rho\rho}(N\pi/d, N'\pi/d, \mathbf{k}_\parallel, t)$, by Eq. (4.8) and using Eqs. (2.3a), (5.22), (6.1), (6.2), and (6.3), the ddcf can be straightforwardly obtained. The singular contributions occur, when $N = N' = 1$ and $|\mathbf{k}_\parallel| \cong k_{\parallel_c}$ and for these wave numbers we find, using that $\Lambda_+^{(0)}(1, k_\parallel \cong k_{\parallel_c}) \cong (\nu + D_T)k_c^2$,

$$M_{\rho\rho}^{(0)}(\pi/d,\pi/d,|\mathbf{k}_\parallel| \cong k_{\parallel_c}, t)$$
$$= M_{\rho\rho}^{(0)}(\pi/d,\pi/d,|\mathbf{k}_\parallel| \cong k_{\parallel_c}, t=0)$$
$$\times \frac{1}{(\nu + D_T)k_c^2}\left\{\nu k_c^2 \exp\left[\frac{-\nu D_T k_c^2 E_>(\mathbf{k}_\parallel)}{(\nu + D_T)}\,t\right] + D_T k_c^2 \exp\left[-k_c^2(\nu + D_T)t\right]\right\}$$
$$+ \frac{\rho k_B T k_{\parallel_c}^2}{(\nu + D_T)^2 k_c^6}\left(\alpha_T \frac{dT_{nc}}{dz_1}\right)^2 \frac{1}{E_>(\mathbf{k}_\parallel)}$$
$$\times\left\{\exp\left[\frac{-\nu D_T k_c^2 E_>(\mathbf{k}_\parallel)}{(\nu + D_T)}\,t\right] - \exp\left[-k_c^2(\nu + D_T)t\right]\right\} \tag{6.4a}$$

where

$$E_>(\mathbf{k}_\parallel) = \frac{4}{3}\,\frac{(k_\parallel - k_{\parallel_c})^2}{k_{\parallel_c}^2} + \left(\frac{R}{R_c} - 1\right) f(\cos\theta) \tag{6.4b}$$

and

$$M_{\rho\rho}^{(0)}(\pi/d, \pi/d, |\mathbf{k}_{\|}| \cong k_{\|_c}, t=0)$$

$$= \frac{\rho^2 k_B T \chi_T(\gamma - 1)}{\gamma} + \frac{\rho k_B T k_{\|_c}^2}{(\nu + D_T) D_T k_c^6} \left(\alpha_T \frac{dT_{nc}}{dz_1} \right)^2 \frac{1}{E_>(\mathbf{k}_{\|})} \tag{6.4c}$$

Equation (6.4c) gives the singular Fourier components of the equal time correlation function $M^{(0)}(\mathbf{R}_1, \mathbf{R}_2)$ and $f(\cos\theta)$ is given by Eq. (5.10c). Like Eqs. (4.11), Eqs. (6.4) have been written in a form valid for all densities, as follows from a hydrodynamic rather than a kinetic treatment.

Comparing Eqs. (4.11) and (6.4), we see that the singular Fourier components of the ddcf above and below the instability are identical, to lowest order in ϵ, if one replaces $(1 - R/R_c)$ below the instability point by $(R/R_c - 1)f(\cos\theta)$ above the instability point. Because of this, the remarks following Eq. (4.11) for the correlations below the instability point are also applicable to Eqs. (6.4). In particular, the singular behavior of $M_{\rho\rho}(t=0)$ for $k_{\|} = k_{\|_c}$ and $R \approx R_c$, that is $\sim[(R/R_c - 1)f(\cos\theta)]^{-1}$, is again due to the contribution of two Λ_- modes to G_2.

In the last part of this section the spatial decay of the equal time ddcf will be examined. As for $R < R_c$, we are interested only in long-range correlations when $R \gtrsim R_c$, so that the first term in Eq. (6.4c) can be neglected since it is of short range. The inverse Fourier transform of the second term in Eq. (6.4c) will be denoted by $D_{\rho\rho}^{(0)}(\mathbf{R}_1, \mathbf{R}_2)$ and for $R \gtrsim R_c$ it is given by

$$D_{\rho\rho}^{(0)}(\mathbf{R}_1, \mathbf{R}_2) \cong \rho k_B T \left(\alpha_T \frac{dT_{nc}}{dz_1} \right)^2 \frac{2}{d} \sin(\pi z_1/d) \sin(\pi z_2/d) \frac{1}{D_T(\nu + D_T)}$$

$$\times \frac{k_{\|_c} d^2}{72\pi^4} \int_0^\infty dk_{\|} \int_0^{2\pi} d\theta$$

$$\times \frac{\exp\left[i k_{\|} R_{12x} \cos\theta + i k_{\|} R_{12y} \sin\theta \right]}{\left[(k_{\|} - k_{\|_c})^2 + (3\pi^2/8d^2)(R/R_c - 1) f(\cos\theta) \right]} \tag{6.5}$$

The asymptotic analysis of Eq. (6.5) for large $|\mathbf{R}_{12_{\|}}|$ is straightforward for two particular cases; case (1) $R_{12y} = 0$ and $R_{12x} \gg d$; case (2) $R_{12x} = 0$ and $R_{12y} \gg d$. Using the method of stationary phase to determine the behavior of Eq. (6.5) for $|\mathbf{R}_{12}| \gg d$, the results for these two limits can be written in the form ($i = 1,2$)

$$D_{\rho\rho}^{(0)}(\mathbf{R}_1,\mathbf{R}_2) \cong \rho k_B T\left(\alpha_T \frac{dT_{nc}}{dz_1}\right)^2 \frac{d\sqrt{6}}{27\pi^3 D_T(\nu + D_T)} \times \sin(\pi z_1/d)\sin(\pi z_2/d) d_i(\mathbf{R}_{12_\parallel}) \qquad (6.6a)$$

where for case (1)

$$d_1 = (R/R_c - 1)^{1/2}\left(R_{12x}/d\sqrt{2}\right)^{-1/2}\cos\left[\pi R_{12x}/d\sqrt{2} - \pi/4\right] \times \exp\left\{-R_{12x}\frac{\pi\sqrt{3}}{2\sqrt{2}d}(R/R_c - 1)^{1/2}\right\} \qquad (6.6b)$$

and for case (2)

$$d_2 = (R/R_c - 1)^{1/2}\left[R_{12y}f(0)/d\sqrt{2}\right]^{-1/2}\cos\left[\pi R_{12y}/d\sqrt{2} - \pi/4\right] \times \exp\left\{-R_{12y}\frac{\pi\left[3f(0)\right]^{1/2}}{2\sqrt{2}\,d}(R/R_c - 1)^{1/2}\right\} \qquad (6.6c)$$

In Eqs. (6.6), for gases ($P \approx 1$), $[f(0)]^{1/2} \approx 0.89$, while for liquids ($P \approx 5$), $[f(0)]^{1/2} \approx 0.675$. We remark that the correlations along the rolls (i.e., d_2) have a longer range and larger amplitude than those across the rolls (i.e., d_1). The similarity of $D_{\rho\rho}^{(0)}$ for $R \gtrsim R_c$ and $D_{\rho\rho}^{s}$ of Eq. (4.14) for $R \lesssim R_c$ shows that the fluctuations in the fluid below the instability point already favor the rolls that will actually appear in the average state only above the instability point. These rolls will more explicitly affect the fluctuations for $R \gtrsim R_c$ if higher order effects in ϵ are taken into account [cf. Section 7(5), Eqs. (7.1)].

In the next section, the results of this paper are reviewed and possible experimental consequences are discussed.

7. DISCUSSION

Here we will discuss in more detail some of the results obtained in the previous sections.

(1) Although we have used in this paper kinetic theory to calculate the density–density correlation functions for dilute gases, our final results have been quoted for general densities, since we have also derived them on the basis of a hydrodynamic theory.[1–4] This hydrodynamic theory, which is valid for all fluid densities, can be found in Ref. 1. It involves coupled hydrodynamic equations for the time evolution of the correlations between

fluctuations of the five conserved quantities δa_α: $M_{\alpha\beta}(\mathbf{R}_1, \mathbf{R}_2, t) \equiv \langle \delta a_\alpha(\mathbf{R}_1, t) \delta a_\beta(\mathbf{R}_2) \rangle_{ss}$, as well as hydrodynamiclike equations for the long-range part of the equal time correlation functions between the δa_α, $D_{\alpha\beta}(\mathbf{R}_1, \mathbf{R}_2)$.[1] Here the δa_α represent the fluctuations of the microscopic mass, momentum, and energy densities from their average values in the steady state.[1]

The solution of these equations can be straightforwardly related to the hydrodynamic eigenvalue problems discussed in Sections 3 and 5, except that in this case the thermodynamic quantities and transport coefficients that appear are those for a general fluid instead of for a dilute gas. The eigenvalue problem can be solved and an approximately complete set of hydrodynamic modes can be used to obtain the $M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2, t)$ results, quoted in this paper. We remark that the hydrodynamic theory is somewhat more elaborate and cumbersome than the kinetic theory. However, the two theories can be developed formally in close parallel and differ only in unimportant technical details. It is therefore not surprising to find a rather trivial difference in the final answers between the two theories, showing that the kinetic theory for dilute gases contains all the essential features also present in a hydrodynamic theory of fluctuations in a dense fluid near a convective instability.

(2) In this paper we have neglected the sound mode contributions to both the equal and unequal time correlation functions. To justify this, we argue as follows. In a previous paper,[3,4] we showed that when k is small and when the walls and gravity are neglected the sound mode contributions to the pair correlation function are very much smaller than the other hydrodynamic mode contributions. Further, the sound mode contributions are not enhanced as R approaches R_c and gravity is included. Therefore, we conclude that the sound mode contributions to the light scattering are not important near $R \cong R_c$. Similar conclusions have been reached before by Lekkerkerker and Boon.[8]

(3) We have shown that the time-dependent correlation functions, in particular the density–density correlation function, exhibit critical slowing down as R approaches R_c from above or below. Physically, this is because a density fluctuation decays via dissipative processes. However, near $R = R_c$ the buoyancy force balances these dissipative forces, so that it takes a density fluctuation an extremely long time to decay to zero.

We have also shown that the equal time correlation functions become extremely long ranged as R approaches R_c from below or above. Although these correlation functions are already long ranged in a nonequilibrium fluid far away from an instability point,[3,4] their range here is of true macroscopic size. Similarly, the length scale of the fluctuations near R_c is quite different from that near a gas–liquid critical point. For, while in the latter two cases the length scale of the equal time correlations is measured in terms of molecular diameters (i.e., close to the critical point the correlation length is many molecular diameters), in the case considered in this paper these correlations are measured in terms of d (i.e., close to R_c the correlation length is a large macroscopic distance).

(4) The theory presented here can—except for the thermal conductivity—be compared with the mean field approximation in critical phenomena. This follows from the fact that both theories are valid in the neighborhood of, but away from, the actual instability or critical point. That is, in both cases there are important corrections to the theory in the immediate vicinity of the singular point. It should be remarked, however, that for the Bénard cell, the region near the instability point, where the calculations of this paper break down, is an experimentally inaccessible region close to R_c (cf. Appendix C). The same is not true for the mean field approximation for critical phenomena.

In Table I we have listed the singular behavior near a gas–liquid critical point, in the mean field approximation, for $T \gtrsim T_c$ and that near the first convective instability in a Bénard cell for $R > R_c$, as given in this paper. We make the following comments on Table I.

(a) We first remark in general that, as is the case near the critical point, the singular behavior (i.e., the critical exponents) above (as quoted in Table I) and below the instability point are the same. We also note that an

Table I. Comparison of Singular Behavior near a Critical and an Instability Point

	T_c	R_c
a. Order parameter	$\rho_L - \rho_G \sim \rho_c\left(\frac{T}{T_c} - 1\right)^{1/2}$	$u_z \sim \frac{D_T}{d}\left(\frac{R}{R_c} - 1\right)^{1/2}$
b. Pair correlation function	$\frac{e^{-R_{12}/\xi}}{(R_{12})^{1/2}}$	$\sim \frac{1}{2(\nu + D_T)} \frac{e^{-R_{12x,y}/\xi_{x,y}}}{(R_{12x,y})^{1/2}} \frac{1}{(R/R_c - 1)^{1/2}}$
c. Correlation length ξ	$\xi \sim \frac{1}{(T/T_c - 1)^{1/2}}$	$\xi_{x,y} \sim \frac{1}{(R/R_c - 1)^{1/2}}$
d. Thermal conductivity λ	$\sim \frac{1}{\nu + D_T} \frac{1}{(T/T_c - 1)^{1/2}}$	$\sim \frac{1}{2(\nu + D_T)} \frac{1}{(R/R_c - 1)^{1/2}}$
e. Critical slowing down	$D_T = \frac{\lambda}{\rho c_p} \sim \frac{1}{\xi} \sim \left(\frac{T}{T_c} - 1\right)^{1/2}$	$\Lambda_-(\mathbf{k}_c) \sim \frac{R}{R_c} - 1$

asymmetric scaling of space is sometimes used[9,22] in the literature and that this scaling leads to the conclusion that $\xi_x \sim (R/R_c - 1)^{-1/2}$ and $\xi_y \sim (R/R_c - 1)^{-1/4}$. This asymmetric scaling in the x and y directions is motivated by examining fluctuations or perturbations in the vicinity of the wavenumber $\mathbf{k}_{\|} \cong k_{\|_c}\hat{\mathbf{x}}$. The thermal fluctuations considered here, however, are not to be restricted to this wavenumber and the results given in Table I are obtained from fluctuations where only the magnitude of $\mathbf{k}_{\|}$ is fixed to be in the vicinity of $k_{\|_c}$.

(b) The expression for u_z follows directly from Eq. (5.1c). It has been experimentally verified both as to amplitude (D_T/d) and exponent $(1/2)$ dependence by Bergé and Dubois.[23]

(c) In three dimensions, the Ornstein–Zernike behavior of $G_2(\mathbf{R}_1, \mathbf{R}_2)$ in equilibrium is $\sim e^{-R_{12}/\xi}/R_{12}$. We have quoted here the two-dimensional result as given by Fisher[24] for the case of large R_{12} at fixed $T > T_c$. It is this behavior which is analogous to that near the instability point in a Bénard cell, because the fluid is of finite extent in the z direction.

For the instability case the importance of mode-coupling has been indicated by a factor $1/2(\nu + D_T)$ and the singular behavior by a factor $(R/R_c - 1)^{-1/2}$. Both come from the contribution of two Λ_- modes in the expression (6.3) for $G_2(\mathbf{R}_1, \mathbf{R}_2)$ [cf. Eq. (5.10b)].

(d) In the instability case there are really three correlation lengths for $R \gtrsim R_c$ cf. Eqs. (6.6b), (6.6c): $\xi_z \sim d$, $\xi_x = (2d\sqrt{2}/\pi\sqrt{3})(R/R_c - 1)^{-1/2}$ and $\xi_y = \xi_x/[f(0)]^{1/2}$.

(e) The importance of mode-coupling in the singular behavior of the thermal conductivity λ near the critical point is shown by the factor $1/(\nu + D_T)$ that indicates a contribution to λ of a viscous and a heat mode. As explained in Appendix B, Eq. (B.7), in the case of the instability, two Λ_- modes, each composed of a viscous and a heat mode, are responsible for the singular behavior of λ near $R = R_c$. However, as shown in Appendix C, the coefficient of the expression for λ near $R = R_c$ given in Table I is so small [in fact $\sim \Gamma$ of Eq. (C.6)] that this singularity cannot be observed.

We note that, although mode-coupling of essentially the same two modes is responsible for the singular behavior of the thermal conductivity near the critical and the instability point, the origin of the singularity is quite different in the two cases. For, near the critical point it is the hydrodynamic eigenfunctions that become singular, involving the singular behavior of the equilibrium pair correlation function, while near the instability point, it is the hydrodynamic eigenvalues, involving the nonequilibrium part of the pair correlation function $\sim (2\Lambda_-)^{-1}$, that become singular.

(f) The critical slowing down near a critical point is determined by two singularities: that of c_p and that of λ. Since $\lambda \sim (T/T_c - 1)^{-1/2}$ and $c_p \sim (T/T_c - 1)^{-1}$, the resulting behavior of D_T is $\sim (T/T_c - 1)^{1/2}$. If one were to ignore the singularity of λ, i.e., in the Van Hove approximation, where the singular behavior of the transport coefficients is neglected with respect to that of the thermodynamic quantities, a behavior of $D_T \sim (T/T_c - 1)$ would be found, analogous to that near the instability. Cummins *et al.*[25] have recently observed that critical slowing down of $\lambda_-(k_z = \pi/d,\ k_\| \cong k_{\|_c})$ for $R < R_c$ by a forced Rayleigh scattering technique. Similarly, the critical slowing down for $R > R_c$, i.e., the vanishing of $\Lambda_-(N = 1,\ k_\| \cong k_{\|_c})$, could be observed.

(5) In Section 6 we computed the equal time density–density correlation function for $R > R_c$ to lowest order in the expansion parameter $\epsilon \sim [R/R_c - 1]^{1/2}$. There it was shown that the correlations are of longer range and have a larger amplitude along than across the rolls. If the perturbation expansion is continued, then in the next order of ϵ we find that although the correlation function along the axis of the rolls, for fixed z, still has essentially the same form as before, the correlation function across the rolls contains additional structure that indicates more explicitly the presence of two-dimensional motion of the fluid in rolls. Denoting the corrections of $O(\epsilon)$ to $D_{\rho\rho}^{(0)}$ [cf. Eq. (6.6a)] by $D_{\rho\rho}^{(1)}$, the explicit results that we have obtained are that

$$\begin{aligned}
&D_{\rho\rho}^{(1)}(R_{12x} > d,\ R_{12y} = 0,\ z_1, z_2, x_1, x_2)\\
&\quad\cong \frac{-\rho k_B T}{D_T(\nu + D_T)}\left(\alpha_T \frac{dT_{nc}}{dz_1}\right)^2 \frac{d}{18\pi^2}\left(\frac{2}{\pi k_{\|_c} R_{12x}}\right)^{1/2}\\
&\qquad\times \left\{\sin(\pi z_2/d)\sin(2\pi z_1/d)\cos\left[k_{\|_c}(R_{12x} + x_1) - \pi/4\right]\right.\\
&\qquad\quad \left. + \sin(\pi z_1/d)\sin(2\pi z_2/d)\cos\left[k_{\|_c}(R_{12x} - x_2) - \pi/4\right]\right\}\\
&\qquad\times \exp\left[-R_{12x}\frac{\pi\sqrt{6}}{4d}(R/R_c - 1)^{1/2}\right]
\end{aligned} \tag{7.1a}$$

and

$$\begin{aligned}
&D_{\rho\rho}^{(1)}(R_{12x} = 0,\ R_{12y} \gg d,\ z_1, z_2,\ x = x_1 = x_2)\\
&\quad\cong \frac{-\rho k_B T}{D_T(\nu + D_T)}\left(\alpha_T \frac{dT_{nc}}{dz_1}\right)^2 \frac{d}{9\pi^2}\,\frac{(50 + 9/P)}{(125 - 27/4)}\left[\frac{2}{\pi k_{\|_c} R_{12y} f(0)}\right]^{1/2}\\
&\qquad\times \cos(k_{\|_c} x)\left(\sin(\pi z_2/d)\sin(2\pi z_1/d) + \sin(\pi z_1/d)\sin(2\pi z_2/d)\right)\\
&\qquad\times \cos\left[k_{\|_c} R_{12y} - \pi/4\right]\exp\left\{-R_{12y}\frac{\pi}{4d}\left[6f(0)\right]^{1/2}(R/R_c - 1)^{1/2}\right\}
\end{aligned} \tag{7.1b}$$

We note that unlike $D_{\rho\rho}^{(0)}$, $D_{\rho\rho}^{(1)}$ is not singular as $(R/R_c - 1) \to 0$. Further, if Eqs. (7.1a) and (7.1b) are expressed in center-of-mass and relative coordinates and if we fix the center-of-mass position, and examine $D_{\rho\rho}^{(1)}$ as a function of $\mathbf{R}_{12_\|}$ then Eq. (7.1a) is seen to oscillate faster than Eq. (7.1b) due to the presence of the rolls, as manifested in the factors $\cos[k_{\|_c}(R_{12x} + x_1) - \pi/4]$ and $\cos[k_{\|_c}(R_{12x} - x_2) - \pi/4]$.

(6) Also of experimental interest are the momentum–momentum, or velocity–velocity correlation functions[14] defined by $(i, y = x, y, z)$

$$\langle \delta p_i(\mathbf{R}_1, t)\delta p_j(\mathbf{R}_2)\rangle_{ss} = M_{p_ip_j}(\mathbf{R}_1, \mathbf{R}_2, t) = m^2 \int d\mathbf{V}_1 \int d\mathbf{V}_2\, V_{1i} V_{2j} C(1t|2) \tag{7.2}$$

Since the calculation of $M_{p_ip_j}(\mathbf{R}_1, \mathbf{R}_2, t)$ is almost identical to the calculations of $M_{\rho\rho}(\mathbf{R}_1, \mathbf{R}_2, t)$ given in Section 4 for $R < R_c$ and in Section 6 for $R \gtrsim R_c$, we will give only the results of these calculations. For $i, j = x, y$ we define the Fourier transform of $M_{p_ip_j}(\mathbf{R}_1, \mathbf{R}_2, t)$ $[\equiv M_{p_ip_j}(k_z, k_z', \mathbf{k}_\|, t)]$ by Eq. (4.8a) with the subscripts $\rho\rho$ replaced by the subscripts p_ip_j and $\sin(k_z z_1)\sin(k_z' z_2)$ replaced by $\cos(k_z z_1)\cos(k_z' z_2)$. If we restrict ourselves to the case where $R \cong R_c$, $k_\| \cong k_{\|_c}$, and $k_z = k_z' = \pi/d$, then for $R \lesssim R_c$ we obtain

$$\begin{aligned} M_{p_ip_j}&(k_z = k_z' = \pi/d, k_\| \cong k_{\|_c}, t) \\ &= \frac{[\delta_{ix}k_y - \delta_{iy}k_x][\delta_{jx}k_y - \delta_{jy}k_x]\rho k_B T \exp[-\nu k_c^2 t]}{k_{\|_c}^2} \\ &\quad + \frac{k_ik_j}{k_{\|_c}^2 k_c^2}\left(\frac{\pi}{d}\right)^2 \frac{\rho k_B T}{(\nu + D_T)} \left\{ \frac{D_T}{E_<(k_\|)} \exp\left[- \frac{k_c^2 \nu D_T E_<(k_\|) t}{(\nu + D_T)} \right] \right. \\ &\qquad \left. + \frac{\nu^2[\nu + 2D_T]}{(\nu + D_T)^2} \exp[-(\nu + D_T)k_c^2 t] \right\} \end{aligned} \tag{7.3}$$

In giving Eq. (7.4), we have retained only the most important contributions for $R \cong R_c$ for each of the modes ν, λ_+, and λ_-. We remark that for $R < R_c$ a more general expression that is valid away from the instability point can be easily derived by using the techniques of Section 4. If $i, j = z$ and if we define $M_{p_zp_z}(k_z, k_z', \mathbf{k}_\|, t)$ by Eq. (4.8a) with $\rho\rho$ replaced by p_zp_z then for $M_{p_zp_z}(k_z = k_z' = \pi/d,\ k_\| \cong k_{\|_c},\ t)$ we obtain Eq. (7.4) with $k_ik_j\pi^2/k_{\|_c}^2 k_c^2 d^2$ replaced by $k_{\|_c}^2/k_c^2$. Finally, for $R \gtrsim R_c$ these correlation functions are given by the same equations except that $E_<(k_\|)$ is replaced by $E_>(\mathbf{k}_\|)$ [cf. Eq. (6.4b)]. The spatial dependence of these correlation functions can be determined in a similar way as was done in the Sections 4

[14] We are indebted for Professors H. L. Swinney and C. Oberman for pointing out to us that these correlation functions could be measured by laser Doppler velocimetry.

and 6 for $M_{\rho\rho}$, using Eq. (4.8b) and leads to similar results [cf. Eqs. (4.14) and (6.6)].

(7) It would be interesting if the singular contributions to the density–density or the momentum–momentum correlation functions could be directly experimentally detected above or below the convective instability. We remark, however, that in order for the critical mode to be probed, the wave number k must be on the order of π/d. If visible light is used to probe such fluctuations, then light scattering techniques at very small angles are required. In fact, if $d \cong 0.1$ cm then the scattering angle must be on the order of 10^{-3} radians. Therefore, as has been remarked before,[8] direct measurement of the density–density correlation functions seems very difficult with visible light. Although indirect measurements of the singular behavior near $R = R_c$ have been reported,[26] it seems to us that a direct measurement, using electromagnetic waves in the microwave regime, would be of considerable interest.

APPENDIX A: THE EIGENMODES FOR $R \gtrsim R_c$

In this appendix, we sketch how the $\Lambda_\pm$ eigenmodes of Eqs. (5.6) with the ansatz (5.9) can be determined.

Inserting Eqs. (5.9) into Eq. (5.6a) and neglecting terms of $O(l/d)$, we obtain the relation

$$n\pi\hat{p}_{jz}(n,m) + ik_y\hat{p}_{jy}(n,m) + i(k_x + ma)\hat{p}_{jx}(n,m) = 0 \tag{A.1a}$$

To the order that the correlation functions, $G_2(12)$ and $C(1t\,|\,2)$, will be calculated we will not need the extension of the viscous mode, ν, to $R \gtrsim R_c$. To compute the extension of the $\lambda_\pm$ modes to $R \gtrsim R_c$, we use that for these modes there is zero vorticity in the z direction, i.e., we require

$$i(k_x + ma)\hat{p}_{jy}(n,m) = ik_y\hat{p}_{jx}(n,m) \tag{A.1b}$$

From Eqs. (A.1b) and (A.1a) we can express $\hat{p}_{jx}$ and $\hat{p}_{jy}$ in terms of $\hat{p}_{jz}$ as

$$\hat{p}_{jx}(n,m) = \frac{n\pi i(k_x + ma)}{\left[k_y^2 + (k_x + ma)^2\right]}\,\hat{p}_{jz}(n,m) \tag{A.1c}$$

$$\hat{p}_{jy}(n,m) = \frac{n\pi ik_y}{\left[k_y^2 + (k_x + ma)^2\right]}\,\hat{p}_{jz}(n,m) \tag{A.1d}$$

where j is the eigenfunction index for the extension of the $\lambda_\pm$ modes to $R \gtrsim R_c$. Next the algebraic equations resulting from Eq. (5.6c) will be given.

Inserting Eqs. (5.9) into Eq. (5.6c), we straightforwardly obtain the following equations for the $\hat{T}_j(n,m)$

$$\begin{aligned}
&\omega_j \hat{T}_j(n,m) - \left[R_c + (R - R_c) \right] \hat{p}_{jz}(n,m) \\
&\quad - \left[k_y^2 + (k_x + ma)^2 + n^2\pi^2 \right] \hat{T}_j(n,m) \\
&= \frac{\epsilon\pi\sqrt{2}}{4} \sum_{\sigma=\pm 1} \sum_{\sigma'=\pm 1} \Big\{ \sigma' a^2 (n - \sigma') \hat{T}_j(n - \sigma', m - \sigma) \\
&\qquad - \sigma a \left[k_x + a(m - \sigma) \right] \hat{T}_j(n - \sigma', m - \sigma) \\
&\qquad - (\pi^2 + a^2) \hat{p}_{jz}(n - \sigma', m - \sigma) \\
&\qquad - \sigma'(\pi^2 + a^2) \frac{i\sigma a}{\pi} \hat{p}_{jz}(n - \sigma', m - \sigma) \Big\} \\
&\quad + \frac{\epsilon^2}{4} (\pi^2 + a^2)^2 a^2 \left[\hat{p}_{jz}(n-2,m) + \hat{p}_{jz}(n+2,m) - \hat{p}_{jz}(2-n,m) \right]
\end{aligned} \tag{A.2}$$

with $\hat{p}_{jz}(s,m) \equiv 0$ for $s < 1$. We remark that in giving Eq. (A.2) we have written $R = R_c + (R - R_c)$ to facilitate an ordering scheme in powers of $\epsilon \sim (R/R_c - 1)^{1/2}$ that will be given later. Further, it is clear from Eq. (A.2) that the coupling between the expansion coefficients with labels (n,m) to expansion coefficients with other labels is proportional to ϵ. This fact will enable us to solve these equations perturbatively in powers of ϵ. Next the algebraic equations that result from Eq. (5.6b) with Eq. (5.9) will be given.

To simplify the algebra, it is convenient to define the differential operator[19]

$$\delta_\alpha = \frac{\partial^2}{\partial R_{1\alpha} \partial R_{1z}} - \delta_{\alpha z} \frac{\partial^2}{\partial R_{1\beta} \partial R_{1\beta}} \tag{A.3a}$$

This operator has the properties

$$\delta_\alpha \frac{\partial}{\partial R_{1\alpha}} = 0 \tag{A.3b}$$

and

$$\begin{aligned}
\delta_\alpha \hat{p}_{j\alpha}(\mathbf{R}_1) &= \frac{\partial}{\partial R_{1z}} \frac{\partial}{\partial R_{1\alpha}} \hat{p}_{j\alpha}(\mathbf{R}_1) - \frac{\partial^2}{\partial R_{1\beta} \partial R_{1\beta}} \hat{p}_{jz}(\mathbf{R}_1) \\
&= - \frac{\partial^2}{\partial R_{1\beta} \partial R_{1\beta}} \hat{p}_{jz}(\mathbf{R}_1)
\end{aligned} \tag{A.3c}$$

Multiplying Eq. (5.6b) by δ_α, using Eqs. (5.9) and (A.3), and performing straightforward but lengthy algebra the resulting equations from Eq. (5.6b) are

$$
\begin{aligned}
&-\omega_j\left[k_y^2+(k_x+ma)^2+n^2\pi^2\right]\hat{p}_{jz}(n,m) \\
&\quad+P\left[k_y^2+(k_x+ma)^2+n^2\pi^2\right]^2\hat{p}_{jz}(n,m) \\
&\quad+P\left[k_y^2+(k_x+ma)^2\right]\hat{T}_j(n,m) \\
&=\frac{-\epsilon\sqrt{2}}{4}\pi \\
&\quad\times\sum_{\sigma=\pm1}\sum_{\sigma'=\pm1}\Bigg\{a^2\left[k_y^2+(k_x+ma)^2\right] \\
&\qquad\times\left[\hat{p}_{jz}(n-\sigma',m-\sigma)+\frac{ia\sigma\sigma'}{\pi}\hat{p}_{jx}(n-\sigma',m-\sigma)\right] \\
&\qquad+\sigma a(k_x+ma)\left[\pi^2 n\sigma'\hat{p}_{jz}(n-\sigma',m-\sigma)\right. \\
&\qquad\qquad\left.+ia\sigma\pi n\hat{p}_{jx}(n-\sigma',m-\sigma)\right] \\
&\qquad+\left[k_y^2+(k_x+ma)^2\right] \\
&\qquad\times\left[\sigma'(n-\sigma')a^2\hat{p}_{jz}(n-\sigma',m-\sigma)\right. \\
&\qquad\qquad\left.-\sigma a(k_x+ma-\sigma a)\hat{p}_{jz}(n-\sigma',m-\sigma)\right] \\
&\qquad-\sigma' a^2\pi n(n-\sigma')i\left[k_y\hat{p}_{jy}(n-\sigma',m-\sigma)\right. \\
&\qquad\qquad\left.+(k_x+ma)\hat{p}_{jx}(n-\sigma',m-\sigma)\right] \\
&\qquad+n\sigma a\pi(k_x+ma-\sigma a)i \\
&\qquad\times\left[k_y\hat{p}_{jy}(n-\sigma',m-\sigma)\right. \\
&\qquad\qquad\left.+(k_x+ma)\hat{p}_{jx}(n-\sigma',m-\sigma)\right]\Bigg\} \qquad \text{(A.4)}
\end{aligned}
$$

Equations (A.1), (A.2), (A.3), and (A.4) form a set of coupled equations for $\hat{p}_{j\alpha}(n,m)$ and $\hat{T}_j(n,m)$ that will now be solved for small values of $\epsilon \sim (R/R_c - 1)^{1/2}$.

The first step in solving these equations is to assume the expansions

$$\begin{aligned} \omega_j &= \omega_j^{(0)} + \epsilon' \omega_j^{(1)} + \epsilon'^2 \omega_j^{(2)} + \cdots \\ \hat{T}_j(n,m) &= \hat{T}_j^{(0)}(n,m) + \epsilon' \hat{T}_j^{(1)}(n,m) + \cdots \\ \hat{p}_{j\alpha}(n,m) &= \hat{p}_{j\alpha}^{(0)}(n,m) + \epsilon' \hat{p}_{j\alpha}^{(1)}(n,m) + \cdots \end{aligned} \tag{A.5}$$

where ϵ' is a formal expansion parameter that characterizes the order of magnitude in ϵ of the terms on the right-hand side of Eq. (A.5) but is set equal to 1 at the end of the calculation. Inserting Eq. (A.5) into Eqs. (A.2) and (A.4), the resulting equations to $O(\epsilon^0)$ are

$$\omega_j^{(0)} \hat{T}_j^{(0)}(n,m) - R_c \hat{p}_{jz}^{(0)}(n,m) - \left[k_y^2 + (k_x + ma)^2 + n^2\pi^2 \right] \hat{T}_j^{(0)}(n,m) = 0 \tag{A.6a}$$

and

$$\omega_j^{(0)} \hat{p}_{jz}^{(0)}(n,m) - P\left[k_y^2 + (k_x + ma)^2 + n^2\pi^2 \right] \hat{p}_{jz}^{(0)}(n,m) - \frac{P\left[k_y^2 + (k_x + ma)^2 \right] \hat{T}_j^{(0)}(n,m)}{\left[k_y^2 + (k_x + ma)^2 + n^2\pi^2 \right]} = 0 \tag{A.6b}$$

Equations (A.6) have two solutions, viz., either $\hat{p}_{jz}^{(0)}(n,m) = 0 = \hat{T}_j^{(0)}(n,m)$ or

$$\left\{ \omega_j^{(0)} \left[k_y^2 + (k_x + ma)^2 + n^2\pi^2 \right] - P\left[k_y^2 + (k_x + ma)^2 + n^2\pi^2 \right]^2 - \frac{P R_c \left[k_y^2 + (k_x + ma)^2 \right]}{\left\{ \omega_j^{(0)} - \left[k_y^2 + (k_x + ma)^2 + n^2\pi^2 \right] \right\}} \right\} = 0 \tag{A.7}$$

To proceed, we first remark that when $\epsilon = 0$ it is clear that $\hat{p}_{jz}(n, m \neq 0) = 0 = \hat{T}_j(n, m \neq 0)$, since the rolls have not yet formed. From this we can then define our perturbation expansion by fixing $\hat{p}_{jz}^{(0)}(n,0)$ and $\hat{T}_j^{(0)}(n,0)$ to be nonzero for some value of n, say N, and requiring all other $p_{jz}^{(0)}$'s and $T_j^{(0)}$'s to be zero. In this way $\omega_j^{(0)}$ can be determined from Eq. (A.7). Thus, we choose

$$\hat{p}_{jz}^{(0)}(N,0) \neq 0 \neq \hat{T}_{jz}^{(0)}(N,0) \tag{A.8a}$$

$\hat{p}_{jz}^{(0)}(n,m) = 0 = \hat{T}_j^{(0)}(n,m)$ for all other (n,m).

From Eq. (A.7) with $n = N$, $m = 0$ we obtain

$$\begin{aligned} \omega_j^{(0)} &\equiv \Lambda_\pm^{(0)}(N,k_\|) \\ &= \frac{(1+P)}{2}\left(k_\|^2 + N^2\pi^2\right) \\ &\quad\times\left\{1 \pm \left[1 - \frac{4P}{(1+P)^2}\left[1 - \frac{R_c k_\|^2}{\left(k_\|^2 + n^2\pi^2\right)^3}\right]\right]^{1/2}\right\} \end{aligned} \tag{A.8b}$$

where $\Lambda_\pm(N,k_\|)$ denotes those eigenvalues that are the extensions of $\lambda_\pm$ to $R \gtrsim R_c$. From Eqs. (A.1), (A.6), and (A.8a), we can obtain the expansion coefficients $\hat{p}_{j\alpha}(N,0)$ and $\hat{T}_j(N,0)$ to $O(\epsilon^0)$ with the results

$$\begin{aligned} \hat{p}_{\Lambda_\pm z}^{(0)}(N,0,\mathbf{k}_\|) &= A_{\Lambda_\pm}(N,k_\|) \\ \hat{T}_{\Lambda_\pm}^{(0)}(N,0,\mathbf{k}_\|) &= \frac{R_c A_{\Lambda_\pm}(N,k_\|)}{\left[\Lambda_\pm^{(0)}(N,k_\|) - \left(k_\|^2 + N^2\pi^2\right)\right]} \\ \hat{p}_{\Lambda_\pm y}^{(0)}(N,0,\mathbf{k}_\|) &= A_{\Lambda_\pm}(N,k_\|)\frac{ik_y}{k_\|^2}N\pi \\ \hat{p}_{\Lambda_\pm x}^{(0)}(N,0,\mathbf{k}_\|) &= A_{\Lambda_\pm}(N,k_\|)\frac{ik_x}{k_\|^2}N\pi \end{aligned} \tag{A.8c}$$

where $A_{\Lambda_\pm}(N,k_\|)$ is a normalizing factor that will be determined later. From Eqs. (A.8), (5.9), (5.5), (5.3b), and (2.11) the unnormalized modes can be easily determined to $O(\epsilon^0)$. We remark, however, that for $N = 1$ and $k_\| \cong k_{\|_c} = \pi/\sqrt{2}\,\Lambda_-^{(0)}$ goes to zero, that is

$$\Lambda_-^{(0)}(1, k_\| \cong k_{\|_c}) \cong \frac{4P}{1+P}\left(k_\| - k_{\|_c}\right)^2 \tag{A.9}$$

Although $\Lambda_-^{(0)}$ for $N > 1$ and $\Lambda_+^{(0)}$ for all N are greater than zero so that they lead to contributions to $C(1t\,|\,2)$ and $G_2(12)$ that are well behaved,[15] this is not true for $\Lambda_-^{(0)}(1, k_\| \cong k_{\|_c})$. Because of this we have to continue

[15] In Section 3, and elsewhere,[1-4] we showed that the long-wavelength contributions to $G_2(12)$ are inversely proportional to the hydrodynamic eigenvalues. From this follows immediately that if one of these eigenvalues goes to zero, $G_2(12)$ is not well behaved.

with the perturbation expansion so that a well-behaved description of the correlation functions for ϵ small but not equal to zero is obtained.

This procedure is straightforward, and since similar calculations can be found in the literature,[21] we quote only the relevant results here. For a general N and $\mathbf{k}_{\|}$ the first-order eigenvalues vanish, i.e.,

$$\Lambda_{\pm}^{(1)}(N, \mathbf{k}_{\|}) = 0 \tag{A.10a}$$

For $\epsilon \neq 0$ a positive definite correction to Eq. (A.9) is given in second-order perturbation theory in ϵ by

$$\begin{aligned}
&\Lambda_{-}^{(2)}\left(N = 1, k_{\|} = \pi/\sqrt{2}\,, \cos\theta\right) \\
&= \frac{\epsilon^2\pi^2}{16(1+P)} \sum_{\sigma=\pm 1} \frac{(1+\sigma\cos\theta)^2}{\left[(5-\sigma\cos\theta)^3 - 27/4(1-\cos\theta)\right]} \\
&\times \left\{ 9\pi^2/2(1-\sigma\cos\theta) + P\pi^2(5-\sigma\cos\theta)^2 \right. \\
&\left. + 3(1-\sigma\cos\theta)\left[\frac{3\pi^2}{2} + \frac{\pi^2}{P}(5-\sigma\cos\theta)\right]\right\}
\end{aligned} \tag{A.10b}$$

The unnormalized right hydrodynamic eigenfunctions that are of interest to us can then be found from Eqs. (A.8), (5.9), (5.5), (5.3b), and (2.1). To lowest order in l/d and ϵ, they are

$$\begin{aligned}
f_l(1)\tilde{\Theta}_{\Lambda_{\pm}}^{R(0)}(1, N, \mathbf{k}_{\|}) &= \frac{e^{i\mathbf{k}_{\|}\cdot R_{1\|}}}{2\pi} \frac{A_{\Lambda_{\pm}}(N, k_{\|})}{m} \phi_l(1) \\
&\times \left\{ \frac{d\log T_{nc}}{dz_1} \frac{\sin(N\pi z_1/d)}{\left[\Lambda_{\pm}^{(0)}(N, k_{\|}) - D_T k^2\right]} \left(\frac{\beta m C_1^2}{2} - \frac{5}{2}\right) \right. \\
&+ \beta m \sin(N\pi z_1/d) C_{1z} \\
&\left. + \beta m \cos(N\pi z_1/d) \frac{iN\pi}{dk_{\|}^2}\left[k_y C_{1y} + k_x C_{1x}\right] \right\}
\end{aligned} \tag{A.11a}$$

where $k^2 \equiv k_{\|}^2 + N^2\pi^2/d^2$. The hydrodynamic eigenvalues to $O(\epsilon^0)$ are

$$\Lambda_{\pm}^{(0)}(N, k_{\|}) = (\nu + D_T)\frac{k^2}{2}\left\{1 \pm \left[1 - \frac{4\nu D_T}{(\nu+D_T)^2}\left(1 - \frac{R_c k_{\|}^2}{d^4 k^6}\right)\right]^{1/2}\right\} \tag{A.11b}$$

Further, if $N = 1$, the critical eigenvalue $\Lambda_-(1, \mathbf{k}_\parallel)$ near $k_\parallel \cong k_{\parallel_c}$ is given to $O(\epsilon^2) \sim (R/R_c - 1)$ by

$$\Lambda_-(1, k_\parallel \cong k_{\parallel_c}, \cos\theta) \cong \frac{\nu D_T k_c^2}{(\nu + D_T)} \left\{ \frac{4}{3} \frac{(k_\parallel - k_{\parallel_c})^2}{k_{\parallel_c}^2} + \left(\frac{R}{R_c} - 1 \right) f(\cos\theta) \right\} \tag{A.11c}$$

with $k_c^2 = 3\pi^2/2d^2$ and $f(\cos\theta)$ given by Eq. (5.10c).

To determine the normalizing factors $A_{\Lambda_\pm}$ in Eq. (A.11a) and to complete the calculation of the hydrodynamic modes for $R \gtrsim R_c$, the adjoint eigenfunctions $\Theta_j^L(1)$ are needed. The hydrodynamic equations for the left eigenvalue problem when $R \gtrsim R_c$ are given by Eq. (2.20) with $n = n_{nc} + \Delta\rho/m$, $T = T_{nc} + \Delta T$ and $\mathbf{u}$ given by Eq. (5.1). This eigenvalue problem can be solved in a manner analogous to that used to determine the right eigenfunctions. Using the results obtained for the left eigenfunctions and the normalization condition

$$\begin{aligned}\left(\Theta_j^L(1, N, \mathbf{k}_\parallel) f_l(1) \Theta_i^R(1, M, \mathbf{k}'_\parallel)\right) &= \left(\Theta_j^{L^{(0)}}(1, N, \mathbf{k}_\parallel) f_l(1) \Theta_i^{R^{(0)}}(1, M, \mathbf{k}'_\parallel)\right) \\ &= \delta_{ij}\delta_{NM}\delta(\mathbf{k}_\parallel - \mathbf{k}'_\parallel)\end{aligned} \tag{A.12}$$

where $i, j = \Lambda_\pm$. The normalized right and left hydrodynamic eigenmodes can then be found to lowest order in ϵ. They are given by the Eqs. (5.11) in the text.

APPENDIX B: SINGULAR CONTRIBUTIONS TO THE HEAT FLUX FOR $R \approx R_c$

In this appendix, we use kinetic theory to calculate mode-coupling contributions to the heat flux in the dilute gas when $R \approx R_c$. These contributions are corrections to the Boltzmann value of the heat flux J_{Bz} in the z direction,

$$J_{Bz} = -\lambda_B \frac{dT}{dz_1} \tag{B.1}$$

where λ_B is the low density (Boltzmann) value of the thermal conductivity. We will outline the theory for $R \lesssim R_c$ and only quote the results for $R \gtrsim R_c$.

The mode-coupling contributions to the thermodynamic fluxes are due to the nonequilibrium pair correlation function, $G_2(12)$, calculated in Section 4 for $R < R_c$. If $G_2(12)$ is retained in the first equation of the BBGKY hierarchy,[3,15,20] then the resulting kinetic equation for the one-particle distribution function in a steady state is

$$\left\{\mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1}+\mathbf{g}\cdot\frac{\partial}{\partial\mathbf{V}_1}\right\}f_1(1)$$
$$=\int d2\,\hat{T}(12)\big[f_1(1)f_1(2)+G_2(12)\big]+\bar{T}_w(1)f_1(1) \qquad \text{(B.2)}$$

In order to obtain a closed equation for $f_1(1)$, we need an expression for $G_2(12)$ in terms of $f_1(1)$. To $O(\mu)$, we can use Eq. (4.2) for $G_2(12)$, which involves $f_\nabla^{(1)}$, and obtain a closed equation for $f_\nabla^{(1)}(1)$. This leads with Eq. (2.3) to an expression for $f_1(1)$. This then implies that for the computation of the heat flux we use the same expression for $G_2(12)$ that led to the singular equal time correlation functions near $R \cong R_c$. Using Eqs. (2.3a) and (4.2) in Eq. (B.2), the resulting equation for $f_\nabla^{(1)}(1)$ is[15]

$$\left\{\mathbf{V}_1\cdot\frac{\partial}{\partial\mathbf{R}_1}+\mathbf{g}\cdot\frac{\partial}{\partial\mathbf{V}_1}\right\}W(\mathbf{R}_1)f_l(1)-W(\mathbf{R}_1)\int d2\,\hat{T}(12)(1+P_{12})f_l(2)f_\nabla^{(1)}(1)$$
$$-\bar{T}_w(1)\big[f_l(1)+f_\nabla^{(1)}(1)\big]$$
$$=\int d2\,\hat{T}(12)\frac{1}{\big[L_{ss}(1)+L_{ss}(2)-\bar{T}_w(1)-\bar{T}_w(2)\big]}$$
$$\times\hat{T}(12)(1+P_{12})W(\mathbf{R}_1)W(\mathbf{R}_2)f_l(2)f_\nabla^{(1)}(1) \qquad \text{(B.3)}$$

Equation (B.3) for $f_\nabla^{(1)}(1)$ takes into account uncorrelated sequences of binary collisions on the left-hand side as given by the Boltzmann equation and a correction to this, viz., a set of correlated binary collisions—called ring events—on the right-hand side.[15,20]

From Eq. (B.3), the heat flux in the z direction can be determined. The result to $O(\mu)$ is[13,15]

$$J_z=k_BT\int d\mathbf{V}_1\,V_{1z}\left(\frac{\beta mV_1^2}{2}-\frac{5}{2}\right)f_\nabla^{(1)}(1)$$
$$=-\lambda_B\frac{dT}{dz_1}-k_BT\int d\mathbf{V}_1\,V_{1z}\left(\frac{\beta mV_1^2}{2}-\frac{5}{2}\right)\frac{1}{\bar{\Lambda}_l(1)}\int d2\,\hat{T}(12)$$
$$\times\frac{1}{\big[L_{ss}(1)+L_{ss}(2)-\bar{T}_w(1)-\bar{T}_w(2)\big]}$$
$$\times\hat{T}(12)(1+P_{12})W(\mathbf{R}_1)W(\mathbf{R}_2)f_l(2)\frac{1}{\bar{\Lambda}_l(1)}$$
$$\times\frac{d\log T}{dz_1}f_l(1)V_{1z}\left(\frac{\beta mV_1^2}{2}-\frac{5}{2}\right)\equiv-\lambda_B\frac{dT}{dz_1}+J_{zR} \qquad \text{(B.4)}$$

with $\mathbf{R}_1$ inside the fluid volume and J_{zR} the ring contribution to J_z.

Using the hydrodynamic modes given by Eq. (3.10), it is straightforward to obtain the long-wavelength contribution of the ring collision events to J_{zR}. Denoting this long-wavelength or hydrodynamic contribution to J_{zR} as J_{zR}^H, we find

$$J_{zR}^H(z_1) = -\frac{5}{2}\frac{k_B}{m}\frac{k_B T}{(\nu + D_T)}\frac{2}{d}\sum_{k_z}\sin^2(k_z z_1)$$
$$\times \int \frac{d\mathbf{k}_\|}{(2\pi)^2}\frac{k_\|^2}{\left(k_\|^2 + k_z^2\right)^2}\frac{1}{\left[1 - R(k_z, k_\|)/R_c\right]}\frac{dT}{dz_1} \tag{B.5}$$

The singular part of J_{zR}^H as $R \to R_c$ is due to the $k_z = \pi/d$ ($n = 1$) contribution in Eq. (B.5). Denoting this singular contribution as J_{zR}^{Hs} yields

$$J_{zR}^{Hs}(z_1) = -\frac{5}{2}\frac{k_B}{m}\frac{k_B T}{(\nu + D_T)}\frac{dT}{dz_1}\frac{2}{d}\sin^2(\pi z_1/d)$$
$$\times \int_0^\infty \frac{dk_\|}{2\pi}\frac{k_\|^2}{\left(k_z^2 + k_\|^2\right)^2}\frac{1}{\left[(1 - R/R_c) + (4/3)(k_\| - k_{\|_c})^2/k_{\|_c}^2\right]} \tag{B.6}$$

where in giving Eq. (B.6) we have used Eq. (4.10a) which is valid approximation as long as R is close to R_c. Using that the integrand in Eq. (B.6) is sharply peaked around $k_\| \cong k_{\|_c}$, $J_{zR}^{Hs}(z_1)$ is easily found to be given to leading order in $(1 - R/R_c)$ by

$$J_{zR}^{Hs}(z_1) = -\frac{c_p k_B T}{(\nu + D_T)}\frac{2}{d}\sin^2(\pi z_1/d)\frac{1}{12\sqrt{3}}\frac{1}{\left[1 - R/R_c\right]^{1/2}}\frac{dT}{dz_1} \tag{B.7}$$

with $c_p = 5k_B/2m$ the specific heat at constant pressure for an ideal gas. In this form Eq. (B.7) is valid for all densities as can be verified by using a hydrodynamic theory. From Eq. (B.7) follows immediately that there is a contribution λ_R^s to the thermal conductivity λ, due to ring events, that reads

$$\lambda_R^s = c_p k_B T\frac{2}{d}\frac{\sin^2(\pi z_1/d)}{(\nu + D_T)}\frac{1}{12\sqrt{3}}\frac{1}{\left[1 - R/R_c\right]^{1/2}} \tag{B.8}$$

and is singular for $R = R_c$.

A similar calculation for $R > R_c$ leads to the same equation for λ_R^s except that $(1 - R/R_c)^{1/2}$ is replaced by

$$(R/R_c - 1)^{1/2}\left\{\left(\int_0^{2\pi} d\theta/\left[f(\cos\theta)\right]^{1/2}\right)\Big/2\pi\right\}^{-1} \cong (R/R_c - 1)^{1/2} \tag{B.9}$$

where $f(\cos\theta)$ is defined by Eq. (5.10c).

APPENDIX C: RENORMALIZATION OF THE CRITICAL EIGENVALUE

The singular ring contribution to the heat flux discussed in Appendix B modifies the hydrodynamic eigenvalues, since the temperature field that should actually be used in the solution of the eigenvalue problem is not determined by Eq. (3.1c) but instead by an equation that takes into account the ring contribution to the heat flux as well. From Eq. (B.4) it follows that this improved equation is

$$-\frac{d}{dz_1}\lambda_B(T)\frac{dT}{dz_1}+\frac{d}{dz_1}J_{zR}(z_1)=0 \tag{C.1}$$

We remark that in determining $J_{zR}^{Hs}(z_1)$, as given by Eq. (B.7), it was assumed that $T(z_1)$ was determined by Eq. (3.1c). Because of this J_{zR}^{Hs} is only the first term in an expansion of the true value of the heat flux in terms of ring collision events.

To solve Eq. (C.1) for the temperature field $T(z_1)$, we formally expand $T(z_1)$ in powers of ring collision events:

$$T(z_1)=T_B(z_1)+\delta T_1(z_1)+\delta^2 T_2(z_1)+\cdots \tag{C.2}$$

Here δ is a formal expansion parameter that only characterizes the order of magnitude of the terms on the right-hand side of Eq. (C.2) and that is set equal to 1 at the end of the calculation, while $T_B(z_1)$ is the Boltzmann value of $T(z_1)$ as determined by Eq. (3.1c). Later δ will be identified with a physical expansion parameter that is small everywhere except in the extreme vicinity of $R \cong R_c$. We now determine $T(z_1)$ to $O(\delta)$.

Inserting Eq. (C.2) into Eq. (C.1), we obtain Eq. (3.1c) to $O(\delta^0)$. Further, retaining only the singular part of J_{zR}, given by Eq. (B.7) to $O(\delta)$, Eq. (C.1) to $O(\delta)$ is

$$\frac{d^2}{dz_1^2}(\lambda_B[T_B]T_1(z_1)) = -\frac{d}{dz_1}\frac{c_p k_B T_B}{(\nu+D_T)}\frac{2}{d}\sin^2(\pi z_1/d)\frac{1}{12\sqrt{3}\,(1-R/R_c)^{1/2}}\frac{dT_B}{dz_1} \tag{C.3}$$

Neglecting the spatial variations of the hydrodynamic quantities on the right-hand side of Eq. (C.3), since they lead to corrections of $O(d/L_\nabla)$, Eq. (C.3) can be straightforwardly solved with the result

$$T_1(z)=\frac{d}{4\pi}\sin(2\pi z_1/d)\frac{2}{d}\frac{k_B T_B}{\rho D_T(\nu+D_T)}\frac{1}{12\sqrt{3}}\frac{1}{(1-R/R_c)^{1/2}}\frac{dT_B}{dz_1} \tag{C.4}$$

where we have used that $T_B(z_1)$ satisfies the boundary conditions at $z_1 = 0$ and d, so that $T_1(z_1 = 0, d) = 0$. To determine the modified hydrodynamic eigenvalues to $O(\delta)$, we need $d[T_B(z_1) + T_1(z_1)]/dz_1$. From Eqs. (C.2) and (C.4) we obtain (again neglecting corrections of $O(d/L_\nabla)$)

$$\frac{d}{dz_1}\left[T_B(z_1) + T_1(z_1)\right] = \left[1 + \frac{\Gamma}{(1 - R/R_c)^{1/2}}\cos(2\pi z_1/d)\right]\frac{dT_B}{dz_1} \tag{C.5}$$

with

$$\Gamma = \frac{k_B T}{d\rho D_T(\nu + D_T)12\sqrt{3}} \tag{C.6}$$

For a gas at STP with $d \sim 0.1$ cm, $\Gamma \sim 10^{-8}$ which is the small quantity related to δ that was mentioned earlier.

To estimate the effect of this correction to dT_B/dz_1 on the eigenvalue problem, we replace $dT/dz_1 = dT_B/dz_1$ on the right-hand side of Eq. (3.3d) by Eq. (C.5) and calculate the modifications to the eigenvalue $\lambda_-(k_z = \pi/d, k_\parallel)$.[16] The resulting eigenvalue problem can be solved without difficulty near $R \cong R_c$ and $k_\parallel \cong k_{\parallel_c}$ by expanding in powers of Γ. The critical eigenvalue is then

$$\lambda_-^R(k_z = \pi/d, k_\parallel \cong k_{\parallel_c}) = \frac{k_c^2 \nu D_T}{(\nu + D_T)}\left[(1 - R/R_c) + \frac{\Gamma}{2(1 - R/R_c)^{1/2}} + \frac{4(k_\parallel - k_{\parallel_c})^2}{3k_{\parallel_c}^2}\right] \tag{C.7}$$

where λ_-^R denotes the renormalized value of λ_- due to the contribution of the ring collision events to J_z. From Eq. (C.7) it follows that the terms $\sim\Gamma$ become important when $(1 - R/R_c) \lesssim \Gamma/2(1 - R/R_c)^{1/2}$ or $(1 - R/R_c) \lesssim \Gamma^{2/3}$. Since $\Gamma \sim 10^{-8}$, the mode-coupling or ring corrections to λ_- are significant only when $(1 - R/R_c) \sim 10^{-6}$. Therefore, unless one is extremely close to the instability point, corrections to the theory presented here are completely insignificant. This means, in the language used in critical phenomena, that a mean field theory of the instability is adequate for all practical purposes. This has been argued before by Graham[(9)] and others[(10,11)] on the basis of fluctuating nonlinear hydrodynamics.

[16] When this ring contribution to the temperature field is included in the kinetic eigenvalue problem, then the basic kinetic streaming operator $L_{ss}(1)$, defined in Section 2, should, for consistency, be modified to include ring collision events. However, one finds that this modification of $L_{ss}(1)$ leads to corrections of $O(d/L_\nabla)$ to the results obtained in Appendix C.

In the last part of the appendix, we argue that the assumption of a fluid with infinite horizontal extent is not realistic for $(1 - R/R_c) \lesssim 10^{-6}$.

From Eq. (4.14) it follows that a correlation length in the horizontal plane, ξ, can be defined

$$\xi = \frac{2\sqrt{2}}{\sqrt{3}} \frac{d}{\pi(1 - R/R_c)^{1/2}} \tag{C.8}$$

Now in an actual experiment a measure of the influence of the horizontal boundaries is the aspect ratio $a = L/d$, where L is the linear dimension of the fluid in the horizontal direction. Typically, the largest aspect ratios used experimentally are $a \sim 50$. Since for $(1 - R/R_c) \sim 10^{-6}$, $\xi/d \sim 10^3$, it follows that the correlation length in the horizontal plane is very much larger than the size of the system, so that the presence of horizontal boundaries can no longer be neglected, as was done in all our calculations here.

REFERENCES

1. T. R. Kirkpatrick, E. G. D. Cohen, and J. R. Dorfman, *Phys. Rev. A* **26**:950 (1982).
2. T. R. Kirkpatrick, E. G. D. Cohen, and J. R. Dorfman, *Phys. Rev. A* **26**:972 (1982).
3. T. R. Kirkpatrick, E. G. D. Cohen, and J. R. Dorfman, *Phys. Rev. A* **26**:995 (1982).
4. T. R. Kirkpatrick, thesis, The Rockefeller University, New York (1981).
5. S. Chandrasekar, *Hydrodynamic and Hydromagnetic Stability* (Clarendon, Oxford, England, 1961).
6. V. Zaitsev and M. Shliomis, *Sov. Phys. JETP* **32**:866 (1971).
7. V. Lesnikov and I. Fisher, *Sov. Phys. JETP* **40**:667 (1975).
8. H. Lekkerkerker and J. Boon, *Phys. Rev. A* **10**:1355 (1974).
9. R. Graham, *Phys. Rev. A* **10**:1762 (1974).
10. R. Graham and H. Pleiner, *Phys. Fluids* **18**:130 (1075).
11. J. Swift and P. C. Hohenberg, *Phys. Rev. A* **15**:319 (1977).
12. Y. Pomeau and P. Résibois, *Phys. Rep.* **19C**:63 (1975).
13. M. Ernst and E. G. D. Cohen, *J. Stat. Phys.* **25**:153 (1981).
14. J. A. Krommes and C. Oberman, *J. Plasma Phys.* **16**:193 (1976).
15. M. Ernst and J. R. Dorfman, *Physica* **61**:157 (1972).
16. J. R. Dorfman and H. van Beijeren, in *Statistical Mechanics, Part B*, B. Berne, ed. (Plenum, New York, 1977), p. 79.
17. T. R. Kirkpatrick and E. G. D. Cohen, *Phys. Lett.* **88A**:44 (1982).
18. J. Niewoudt, thesis, University of Maryland (1982).
19. A. Schlüter, P. Lortz, and F. Busse, *J. Fluid Mech.* **23**:129 (1965).
20. J. R. Dorfman and E. G. D. Cohen, *Phys. Lett.* **16**:124 (1965).
21. D. D. Joseph, *Stability of Fluid Motions, II* (Springer-Verlag, Berlin, 1976).
22. See, for example, A. C. Newell and J. A. Whitehead, *J. Fluid Mech.* **38**:279 (1969) and M. C. Cross, *Phys. Rev. A* **25**:1065 (1982).
23. P. Bergé and M. Dubois, *Phys. Rev. Lett.* **32**:1041 (1974).
24. M. Fisher, *J. Math. Phys.* **5**:957 (1964).
25. C. Allain, H. Z. Cummins, and P. Lallemand, *J. Phys. Lett. (Paris)* **39**:473 (1978).
26. M. Sano and Y. Sawada, *Prog. Theor. Phys. Suppl.* **64**:202 (1978).