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Nonanalytic Dispersion Relations for Classical Fluids. II. The General Fluid

M. H. Ernst[1] and J. R. Dorfman[2]

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The analytic structure of the hydrodynamic frequencies $z(k)$ for the sound, heat, and shear modes and of the hydrodynamic equations for a monatomic fluid are discussed on the basis of the mode–mode coupling theory. It is shown that the hydrodynamic frequencies depend on the wave number k, for small k, as $z(k) = ak + bk^2 + \sum_{n=1}^{\infty} c_n k^{3-2^{-n}}$, and that some of the correlation functions that appear in the Fourier-Laplace transforms of the hydrodynamic equations contain branch point singularities. The implications of these results for the derivation of linear hydrodynamic equations, such as the Burnett equations, and for the long-time behavior of time correlation functions are discussed.

KEY WORDS: Statistical mechanics; nonequilibrium statistical mechanics; hydrodynamic equations; Navier–Stokes equations; Burnett equations; time correlation functions.

1. INTRODUCTION

The study of nonanalytic dispersion relations in classical fluids was begun in a previous paper[(1)], hereafter referred to as I. In I we discussed kinetic theory for a moderately dense gas of hard spheres, and showed that as a

[1] Institute for Theoretical Physics, University of Utrecht, The Netherlands.
[2] Institute for Fluid Dynamics and Applied Mathematics, and Department of Physics and Astronomy, University of Maryland, College Park, Maryland.

result of certain dynamical events taking place in the gas, hydrodynamic frequencies, such as the sound mode frequency z_σ, have a nonanalytic dependence on the wave number k of the form

$$z_\sigma = \pm ick + ak^2 + bk^{5/2} + \cdots \tag{1}$$

We arrived at this result by deriving what are known as the mode–mode coupling integrals on the basis of dynamical arguments, and expressing the hydrodynamic frequencies directly in terms of these integrals.

To discuss the hydrodynamic frequencies for a more general class of fluids by arguments similar to those in I, a fundamental dynamical theory would be needed for such fluids. At the present time such a theory is not available. Instead, we shall assume that for a general class of fluids the hydrodynamic frequencies are determined by the mode–mode coupling theory formulated on the basis of phenomenological arguments by Kadanoff and Swift,[2] Kawasaki,[3] and Ferrell,[4] and on the basis of somewhat different arguments by Ernst *et al.*[5] Using the mode–mode coupling theory, we show that the nonanalytic wave number dependence typified by Eq. (1) also holds for a general class of fluids, and recover the results of I in the limit of low density. In addition, we verify a conclusion of Pomeau's[6] that there is, in the dispersion relation for the hydrodynamic frequencies, an infinite number of powers of k between k^2 and k^3 with the general form k^{2+P_n}, where $P_n = 1 - 2^{-n}$.

We will discuss in some detail other analytic features of the decay of hydrodynamic disturbances in fluids that result from the mode–mode coupling theory, either as given here or as derived in I. These are: (i) the divergence of the transport coefficients in the Burnett and super-Burnett hydrodynamic equations which result if one assumes *ab initio* that the corrections to the Navier–Stokes equations may be expanded in powers of the wave number, or in higher order gradients of the hydrodynamic variables; (ii) the presence of branch point singularities in the mode–mode integrals, noted first by Dufty[7]; and (iii) the relation between the k^{2+P_n} terms in the dispersion relation and the $t^{-(1+P_n)}$ terms which appear in the long-time behavior of the time correlation functions that determine the Navier–Stokes transport coefficients.

The plan of the paper is as follows. In Section 2 we will give a derivation of the hydrodynamic equations in a form most useful for our discussion. In Section 3 we apply the mode–mode theory and obtain the dispersion relations. In Section 4 we discuss the effect of the branch point singularities on the decay of hydrodynamic disturbances. In Section 5 we consider the long-time decay of the time correlation functions, and also discuss the convergence of the infinite series of terms that appears in the dispersion relations. In Section 6 we discuss the divergence of the transport coefficients associated with the

Burnett and super-Burnett equations. Finally, in Section 7 we discuss our results.

2. GENERALIZED HYDRODYNAMICS

Consider a collection of N identical particles of mass m, confined to a volume V. The particles interact with central, pairwise forces and obey classical mechanics. The hydrodynamic equations for such a system are equations for the average number, momentum, and energy densities, which are considered to be suitably defined averages of the microscopic number density $n(\mathbf{r})$, momentum density $\mathbf{g}(\mathbf{r})$, and energy density $e(\mathbf{r})$:

$$\begin{aligned} n(\mathbf{r}) &= \sum_{i=1}^{N} \delta(\mathbf{r}_i - \mathbf{r}) \\ \mathbf{g}(\mathbf{r}) &= \sum_{i=1}^{N} m\mathbf{v}_i\, \delta(\mathbf{r}_i - \mathbf{r}) \\ e(\mathbf{r}) &= \sum_{i=1}^{N} e_i\, \delta(\mathbf{r}_i - \mathbf{r}) \end{aligned} \tag{2}$$

Here $\mathbf{r}_i$ and $\mathbf{v}_i$ denote the position and velocity of particle i, and e_i is the energy of the ith particle:

$$e_i = \tfrac{1}{2}mv_i^2 + \frac{1}{2}\sum_{j(\neq i)} \phi(r_{ij}) \tag{2a}$$

where $\phi(r)$ is the pair potential and $r_{ij} = |\mathbf{r}_{ij}| = |\mathbf{r}_i - \mathbf{r}_j|$, so that $H = \int d\mathbf{r}\, e(\mathbf{r})$ is the Hamiltonian of the system.

The average number, momentum, and energy densities that appear in the hydrodynamic equations are denoted by $\langle a(\mathbf{r}, t)\rangle$, where $a = n$, $\mathbf{g}$, or e. The nonequilibrium average is taken over the normalized initial distribution function $\rho_0(1 + \Phi_N(0))$, where ρ_0 is the grand canonical equilibrium distribution function

$$\rho_0 = (Z_{\mathrm{gr}}N!)^{-1}(m/h)^{3N}\exp(-\beta H + \nu N) \tag{3}$$

Here h is Planck's constant; $\beta = 1/k_{\mathrm{B}}T$, where k_{B} is Boltzmann's constant and T is temperature; $\nu = \beta\mu$, where μ is the chemical potential; and Z_{gr} is the grand canonical partition function

$$Z_{\mathrm{gr}} = \sum_{N} (N!)^{-1}(m/h)^{3N}\int d\Gamma_N \exp(-\beta H + \nu N) \tag{4}$$

where $d\Gamma_N = d\mathbf{r}_1\, d\mathbf{r}_2 \cdots d\mathbf{r}_N\, d\mathbf{v}_1\, d\mathbf{v}_2 \cdots d\mathbf{v}_N$ is the differential volume in phase space. The mass m appears in (3) and (4) since we have chosen the velocities instead of the momenta as phase space variables. The quantity $\Phi_N(0)$ represents the initial deviation from total equilibrium, and since the initial distribution function and ρ_0 are normalized, we have

$$\sum_N (N!)^{-1}(m/h)^{3N} \int d\Gamma_N \, \rho_0 \Phi_N(0) = 0 \tag{5}$$

The nonequilibrium average is now given by

$$\langle a(\mathbf{r}, t)\rangle = \langle a(\mathbf{r}, t)\rangle_{\text{eq}} + \langle \Phi_N(0) a(\mathbf{r}, t)\rangle_{\text{eq}} \tag{6}$$

where $\langle \cdots \rangle_{\text{eq}}$ is an average over the grand canonical distribution (3). The time dependence of the microscopic function $a(\mathbf{r}, t)$ is formally given by

$$a(\mathbf{r}, t) = e^{tL} a(\mathbf{r}) \tag{7}$$

where L is the Liouville operator, defined as

$$L \cdots = \{\cdots, H\} \tag{8}$$

and $\{\cdots, \cdots\}$ are the Poisson brackets. It will be convenient to impose periodic boundary conditions and to consider Fourier transforms of the microscopic densities, defined in general as

$$a_{\mathbf{k}} = \int_V d\mathbf{r} \, [\exp(-i\mathbf{k}\cdot\mathbf{r})]\{a(\mathbf{r}) - \langle a(\mathbf{r})\rangle_{\text{eq}}\} \tag{9}$$

so that due to translational invariance

$$\begin{aligned} n_{\mathbf{k}} &= \sum_{i=1}^{N} \exp(-i\mathbf{k}\cdot\mathbf{r}_i) - V\langle n(\mathbf{r})\rangle_{\text{eq}} \, \delta_{\mathbf{k},0} \\ \mathbf{g}_{\mathbf{k}} &= \sum_{i=1}^{N} m\mathbf{v}_i \exp(-i\mathbf{k}\cdot\mathbf{r}_i) \\ e_{\mathbf{k}} &= \sum_{i=1}^{N} e_i[\exp(-i\mathbf{k}\cdot\mathbf{r}_i)] - V\langle e(\mathbf{r})\rangle_{\text{eq}} \, \delta_{\mathbf{k},0} \end{aligned} \tag{10}$$

where $\delta_{\mathbf{k},0}$ is a Kronecker delta, and $\langle n(\mathbf{r})\rangle_{\text{eq}} = n$ and $\langle e(\mathbf{r})\rangle_{\text{eq}} = e$ are, respectively, the average number and energy density at thermal equilibrium. For $k = 0$ we have

$$n_0 = N - \langle N\rangle_{\text{eq}}, \qquad \mathbf{g}_0 = \mathbf{P}, \qquad e_0 = H - \langle H\rangle_{\text{eq}} \tag{10a}$$

where $\mathbf{P}$ is the total momentum of the system. In the sequel we will also need Laplace transforms of microscopic densities, defined as

$$a_{\mathbf{k}z} = \int_0^\infty dt \, e^{-zt} a_{\mathbf{k}}(t) = \mathscr{G}_z a_{\mathbf{k}} \tag{11}$$

where $\mathscr{G}_z = (z - L)^{-1}$ is the resolvent operator, and according to (6) and (9), the average $\langle a_{\mathbf{k}z}\rangle$ is given by

$$\langle a_{\mathbf{k}z}\rangle = \sum_N (Z_{\mathrm{gr}}N!)^{-1}(m/h)^{3N}\int d\Gamma_N\, \rho_0\Phi_N(0)(z - L)^{-1}a_{\mathbf{k}} \tag{12}$$

The macroscopic quantities of interest to us, $\langle a(\mathbf{r}, t)\rangle$, can therefore be determined in terms of the inverse Fourier–Laplace transforms of the averages of $a_{\mathbf{k}z}$, and we now turn our attention to the development of the Zwanzig projection operator method[8] for obtaining the generalized hydrodynamic equations satisfied by $\langle a_{\mathbf{k}z}\rangle$. We first introduce an inner product between microscopic variables, such as $a_{\mathbf{k}}$ and $b_{\mathbf{q}}$, as

$$(a_{\mathbf{k}}, b_{\mathbf{q}}) = V^{-1}\langle a_{\mathbf{k}}{}^*b_{\mathbf{q}}\rangle_{\mathrm{eq}} \tag{13}$$

where the asterisk denotes complex conjugation. Then, because of the translational invariance of the equilibrium average, the inner product satisfies

$$(a_{\mathbf{k}}, b_{\mathbf{q}}) = \delta_{\mathbf{k},\mathbf{q}}(a_{\mathbf{k}}, b_{\mathbf{k}}) \tag{14}$$

Instead of using the microscopic variables $n_{\mathbf{k}}$ and $e_{\mathbf{k}}$ it is more convenient to use linear combinations representing the local pressure $P_{\mathbf{k}}$ and the entropy per particle $S_{\mathbf{k}}$:

$$P_{\mathbf{k}} = (\partial P/\partial e)_n e_{\mathbf{k}} + (\partial P/\partial n)_e n_{\mathbf{k}}, \qquad S_{\mathbf{k}} = (nT)^{-1}\{e_{\mathbf{k}} - hn_{\mathbf{k}}\} \tag{15}$$

Here $h = n^{-1}(e + P)$ and P is the equilibrium pressure. From the quantities S_k, $P_{\mathbf{k}}$, and $\mathbf{g}_{\mathbf{k}}$ we construct an approximately orthonormal set $a_{\mathbf{k}}{}^i$ with

$$(a_{\mathbf{k}}{}^i, a_{\mathbf{k}}{}^j) = \delta_{ij} + O(k^2) \tag{16}$$

where the labels (i, j) stand for T, $\sigma = \pm$, η_1, or η_2, with

$$a_{\mathbf{k}}{}^T = (n/k_{\mathrm{B}}C_P)^{1/2}S_{\mathbf{k}} \tag{17a}$$

$$a_{\mathbf{k}}{}^\sigma = (\beta/2\rho)^{1/2}\{c^{-1}P_{\mathbf{k}} + \sigma\hat{\mathbf{k}}\cdot\mathbf{g}_{\mathbf{k}}\} \tag{17b}$$

$$a_{\mathbf{k}}^{\eta_i} = (\beta/\rho)^{1/2}\hat{\mathbf{k}}^i\cdot\mathbf{g}_{\mathbf{k}} \tag{17c}$$

The set of unit vectors $\hat{\mathbf{k}}$, $\hat{\mathbf{k}}_\perp{}^1$, and $\hat{\mathbf{k}}_\perp{}^2$ are mutually orthogonal. The mass density $\rho = mn$, the specific heat per particle at constant pressure is C_P, and the zero-frequency adiabatic sound velocity is c, defined by $c^2 = (\partial P/\partial\rho)_S$. These microscopic variables are closely related to the hydrodynamic modes of the kinetic equation used in I. The orthogonality condition to order k^2 in (16) will be sufficient here. In order to verify the relations (16) one needs a number of fluctuation formulas. These formulas, and others needed for later developments in this paper, are discussed in Appendix A. In fact, one can make the orthonormality conditions (16) exact to all orders in k by introducing k-dependent generalizations of the thermodynamic quantities $(\partial P/\partial e)_n$,

$(\partial P/\partial n)_e$, and h in (15). This introduces k-dependent generalizations of the specific heat C_P and the sound velocity c.[2,9,10] For small values of k, all k-dependent quantities deviate from their lowest order contribution by terms of relative order k^2, whose coefficients can be expressed in terms of equilibrium distribution functions.

After this introduction on notation and definitions we turn our attention to the equations of generalized hydrodynamics. The derivation of these equations also yields explicit expressions for the generalized transport coefficients, which will be needed for actual calculations. Therefore, a projection operator P is defined as

$$Pb = \sum_{\mathbf{k}} \sum_{i} a_{\mathbf{k}}^{i}(a_{\mathbf{k}}^{i}, b) \tag{18}$$

where the sum over i designates the five functions given by (17a)–(17c), and b is an arbitrary function of the phase variables. We also define $P_\perp = 1 - P$, and notice that $P^2 = P$ as a consequence of (16). When the projection operator P acts on a special Fourier component $b_{\mathbf{k}}$, it reduces to

$$Pb_{\mathbf{k}} = \sum_{i} a_{\mathbf{k}}^{i}(a_{\mathbf{k}}^{i}, b_{\mathbf{k}}) \tag{19}$$

by virtue of (14). The operator P projects on the hydrodynamic subspace spanned by $e_{\mathbf{k}}$, $n_{\mathbf{k}}$, and $\mathbf{g}_{\mathbf{k}}$, or by the $a_{\mathbf{k}}^{i}$ in (17a)–(17c), and $P_\perp$ projects on its orthogonal complement.

We now use the projection operator to solve the equation of motion for $a_{\mathbf{k}z}^{r}$, defined in (11), which reads

$$(z - L)a_{\mathbf{k}z}^{r} = a_{\mathbf{k}}^{r} \tag{20}$$

We write $a_{\mathbf{k}z}^{r} = Pa_{\mathbf{k}z}^{r} + P_\perp a_{\mathbf{k}z}^{r}$, apply P and $P_\perp$ respectively to (20), solve the second equation for $P_\perp a_{\mathbf{k}z}^{r}$, and insert the result into the first equation, obtaining

$$[z - PLP - PLP_\perp\{z - P_\perp LP_\perp\}^{-1}P_\perp LP]Pa_{\mathbf{k}z}^{r} = a_{\mathbf{k}}^{r} \tag{21}$$

Written out in component form this equation is

$$\sum_{j} \{z\,\delta_{ij} + ik\Omega_{ij}(\mathbf{k}) + k^2 U_{ij}(\mathbf{k}, z)\}\mathscr{G}_{jr}(\mathbf{k}, z) = \delta_{ir} \tag{22}$$

where $\mathscr{G}_{ij}$ are hydrodynamic propagators or correlation functions of hydrodynamic variables

$$\mathscr{G}_{ij}(\mathbf{k}, z) = (a_{\mathbf{k}}^{i}, a_{\mathbf{k}z}^{j}) = (a_{\mathbf{k}}^{i}, \mathscr{G}_z a_{\mathbf{k}}^{j}) \tag{23}$$

The matrix $\Omega_{ij}(\mathbf{k})$ is referred to as the frequency matrix and is defined by

$$-ik\Omega_{ij}(\mathbf{k}) = (a_{\mathbf{k}}^{i}, La_{\mathbf{k}}^{j}) \tag{24}$$

The transport matrix U_{ij} is defined by

$$-k^2 U_{ij}(\mathbf{k}, z) = (a_{\mathbf{k}}{}^i, LP_{\perp z}\hat{\mathscr{G}}P_{\perp}La_k{}^j) \tag{25}$$

where we have introduced the projected resolvent operator

$$\hat{\mathscr{G}}_z = P_{\perp}(z - P_{\perp}LP_{\perp})^{-1}P_{\perp} \tag{26}$$

Equations (22) for the hydrodynamic propagators are exact. They are, however, not the equations of motion for Fourier–Laplace transforms of the macroscopic hydrodynamic variables, which can be expressed as linear combinations of the quantities $\langle a_{\mathbf{k}z}^i\rangle$ defined in (12) as

$$\langle a_{\mathbf{k}z}^i\rangle = \langle \Phi_N(0)a_{\mathbf{k}z}^i\rangle_{\mathrm{eq}} \tag{27}$$

To find the equation of motion for $\langle a_{\mathbf{k}z}^i\rangle$, we write

$$a_{\mathbf{k}z}^i = Pa_{\mathbf{k}z}^i + P_{\perp}a_{\mathbf{k}z}^i = Pa_{\mathbf{k}z}^i + P_{\perp}\hat{\mathscr{G}}_z P_{\perp}LPa_{\mathbf{k}z}^i \tag{28}$$

where the second equality follows directly from applying $P_{\perp}$ to (20). By inserting (28) into (27) and using (19), we obtain

$$\langle a_{\mathbf{k}z}^i\rangle = \sum_r \mathscr{G}_{ri}(\mathbf{k}, z)\langle \Phi_N(0)\{a_{\mathbf{k}}{}^r + P_{\perp}\hat{\mathscr{G}}_z P_{\perp}La_{\mathbf{k}}{}^r\}\rangle_{\mathrm{eq}} \tag{29}$$

Since $\mathscr{G}_{ri}(\mathbf{k}, z) = \mathscr{G}_{ir}(\mathbf{k}, t)$, as shown below, the generalized hydrodynamic equations can be obtained by multiplying (22) by $\langle \Phi_N(0)\{a_{\mathbf{k}}{}^r + P_{\perp}\hat{\mathscr{G}}_z P_{\perp}La_{\mathbf{k}}{}^r\rangle_{\mathrm{eq}}$ and summing over r, and are given by

$$\sum_j \{z\,\delta_{ij} + ik\Omega_{ij}(\mathbf{k}) + k^2 U_{ij}(\mathbf{k}, z)\}\langle a_{\mathbf{k}z}^j\rangle$$
$$= \langle a_{\mathbf{k}}{}^i(0) + P_{\perp}\hat{\mathscr{G}}_z P_{\perp}La_{\mathbf{k}}{}^i(0)\rangle \tag{30}$$

The second term on the right-hand side of (30) serves as a correction term to the initial condition term $\langle a_{\mathbf{k}}{}^i(0)\rangle$. In the usual form of the linearized hydrodynamic equations, it is not taken into account since it is of order k.

Having obtained equations for the hydrodynamic propagators (22) and the linear generalized hydrodynamic equations (30), we devote the rest of this section to obtaining more explicit expressions for the matrices Ω_{ij} and U_{ij}. First, note that $\mathscr{G}_{ij}(\mathbf{k}, z)$, $\Omega_{ij}(k)$, and $U_{ij}(\mathbf{k}, z)$ are symmetric on interchange of i and j, which can be proved by using Liouville's theorem and the transformation $\mathbf{r}_i \to -\mathbf{r}_i$. These matrices are symmetric because we are using orthonormal basis functions, such as $a_{\mathbf{k}}{}^i$. The advantage of this choice of basis functions (17) is that it diagonalizes the matrix $\mathbf{\Omega}(\mathbf{k})$ to lowest order in k.

The quantities $La_{\mathbf{k}}{}^i$ appearing in the expressions for Ω_{ij} and U_{ij} represent the rate of change of (linear combinations of) the microscopic particle, energy, and momentum densities, and satisfy microscopic conservation laws of the form

$$La_{\mathbf{k}}^{i} = -i\mathbf{k}\cdot\mathbf{j}_{\mathbf{k}}^{i} = -ikj_{\mathbf{k}x}^{i} \tag{31}$$

where, for convenience, we take $\mathbf{k} = k\hat{\mathbf{x}}$, where $\hat{\mathbf{x}}$ is a unit vector in the x direction. The quantities $j_{\mathbf{k}x}^{i}$, from (8) and (17), are

$$j_{\mathbf{k}x}^{T} = (n/k_{\mathrm{B}}C_P)^{1/2}j_{\mathbf{k}x}^{s} = (\beta/nTC_P)^{1/2}(j_{\mathbf{k}x}^{e} - hj_{\mathbf{k}x}^{n}) \tag{32a}$$

$$j_{\mathbf{k}x}^{\sigma} = \frac{1}{\sqrt{2}}\left[\frac{1}{c}\left(\frac{\beta}{\rho}\right)^{1/2} j_{\mathbf{k}x}^{P} + \sigma j_{\mathbf{k}x}^{l}\right]$$

$$= \left(\frac{\beta}{2\rho}\right)^{1/2}\left[\frac{1}{c}\left(\frac{\partial P}{\partial e}\right)_n j_{\mathbf{k}x}^{e} + \frac{1}{c}\left(\frac{\partial P}{\partial n}\right)_e j_{\mathbf{k}x}^{n} + \sigma\tau_{\mathbf{k}xx}\right] \tag{32b}$$

$$j_{\mathbf{k}x}^{l} = (\beta/\rho)^{1/2}\tau_{\mathbf{k}xx} \tag{32c}$$

$$j_{\mathbf{k}x}^{\eta_1} = (\beta/\rho)^{1/2}\tau_{\mathbf{k}xy} \tag{32d}$$

$$j_{\mathbf{k}x}^{\eta_2} = (\beta/\rho)^{1/2}\tau_{\mathbf{k}xz} \tag{32e}$$

where

$$j_{\mathbf{k}x}^{n} = \sum_{i=1}^{N} v_{ix}\exp(-i\mathbf{k}\cdot\mathbf{r}_i) \tag{33a}$$

$$j_{\mathbf{k}x}^{e} = \sum_{i=1}^{N}\left[e_i v_{ix} + \sum_{\alpha=x,y,z}\tau_{i,x\alpha}(\mathbf{k})v_{i\alpha}\right]\exp(-i\mathbf{k}\cdot\mathbf{r}_i) \tag{33b}$$

$$\tau_{\mathbf{k}x\alpha} = \sum_{i=1}^{N}[mv_{ix}v_{i\alpha} + \tau_{i,x\alpha}(\mathbf{k})]\exp(-i\mathbf{k}\cdot\mathbf{r}_i) \tag{33c}$$

with $\alpha = x$, y, or z, and

$$\tau_{i,x\alpha}(\mathbf{k}) = \frac{1}{2}\sum_{j(\neq i)} r_{ij,x}\frac{\partial\phi(r_{ij})}{\partial r_{ij,\alpha}}\,\frac{1-\exp(i\mathbf{k}\cdot\mathbf{r}_{ij})}{i\mathbf{k}\cdot\mathbf{r}_{ij}} \tag{34}$$

Expressions (33a)–(33c) for the particle, energy, and momentum densities are considered previously by Schofield.[9] The frequency matrix can be written as

$$\Omega_{ij}(\mathbf{k}) = (a_{\mathbf{k}}^{i}, j_{\mathbf{k}x}^{j}) = (j_{\mathbf{k}x}^{i}, a_{\mathbf{k}}^{j}) \tag{35}$$

and

$$U_{ij}(\mathbf{k}, z) = (\hat{j}_{\mathbf{k}x}^{i}, \hat{\mathscr{G}}_z\hat{j}_{\mathbf{k}x}^{j}) \tag{36}$$

where we have introduced the projected currents

$$\hat{j}_{\mathbf{k}x}^{i} = P_{\perp}j_{\mathbf{k}x}^{i} = j_{\mathbf{k}x}^{i} - \sum_{j} a_{\mathbf{k}}^{j}\Omega_{ji}(\mathbf{k}) \tag{37}$$

An observation that greatly reduces the number of matrix elements Ω_{ij} and U_{ij} is that all matrix elements of Ω_{ij} and U_{ij} vanish when one index denotes a shear mode η_i and the other index denotes a non-shear mode, or when each index refers to a different shear mode. In either case the required velocity integrals vanish. It is now possible to compute $\Omega_{ij}(k)$, and we obtain

$$\Omega_{\sigma\sigma}(\mathbf{k}) = \sigma c \tag{38a}$$

and

$$\Omega_{ij}(\mathbf{k}) = 0 \tag{38b}$$

in all other cases, where we have used the relations

$$(j_{\mathbf{k}x}^s, g_{\mathbf{k}x}) = (S_\mathbf{k}, \tau_{\mathbf{k}xx}) = 0 \tag{39}$$

$$(j_{\mathbf{k}x}^P, g_{\mathbf{k}x}) = (P_\mathbf{k}, \tau_{\mathbf{k}xx}) = \rho c^2/\beta \tag{40}$$

The first equalities in (39) and (40) follow from (35); the second set of equalities is discussed in Appendix A. We first consider the components of the currents in the hydrodynamic subspace, i.e., $Pj_{\mathbf{k}x}^i$. From (19), (39), and (40), we deduce immediately

$$Pj_{\mathbf{k}x}^n = \rho^{-1}\beta g_{\mathbf{k}x}(g_{\mathbf{k}x}, j_{\mathbf{k}x}^n) = j_{\mathbf{k}x}^n \tag{41a}$$

$$Pj_{\mathbf{k}x}^s = \rho^{-1}\beta g_{\mathbf{k}x}(g_{\mathbf{k}x}, j_{\mathbf{k}x}^s) = 0 \tag{41b}$$

$$P\tau_{\mathbf{k}xx} = (n/k_\mathrm{B}C_P)S_\mathbf{k}(S_\mathbf{k}, \tau_{\mathbf{k}xx}) + (\beta/\rho c^2)P_\mathbf{k}(P_\mathbf{k}, \tau_{\mathbf{k}xx}) = P_\mathbf{k} \tag{41c}$$

$$P\tau_{\mathbf{k}xy} = P\tau_{\mathbf{k}xz} = 0 \tag{41d}$$

The nonhydrodynamic components $\hat{j}_{\mathbf{k}x}^i$ are, according to (37) and (41),

$$\hat{j}_{\mathbf{k}x}^T = (\beta/nTC_P)^{1/2}\hat{j}_{\mathbf{k}x}^e \tag{42a}$$

$$\hat{j}_{\mathbf{k}x}^\sigma = [(\gamma - 1)/2]^{1/2}\hat{j}_{\mathbf{k}x}^T + (\sigma/\sqrt{2})\hat{j}_{\mathbf{k}x}^l \tag{42b}$$

$$\hat{j}_{\mathbf{k}x}^l = (\beta/\rho)^{1/2}\hat{\tau}_{\mathbf{k}xx} \tag{42c}$$

$$\hat{j}_{\mathbf{k}x}^{\eta_1} = (\beta/\rho)^{1/2}\tau_{\mathbf{k}xy} \tag{42d}$$

$$\hat{j}_{\mathbf{k}x}^{\eta_2} = (\beta/\rho)^{1/2}\tau_{\mathbf{k}xz} \tag{42e}$$

where

$$\hat{j}_{\mathbf{k}x}^e = j_{\mathbf{k}x}^e - hj_{\mathbf{k}x}^n \tag{43a}$$

$$\hat{\tau}_{\mathbf{k}xx} = \tau_{\mathbf{k}xx} - P_\mathbf{k} \tag{43b}$$

To derive (42b), we need the thermodynamic identity $(TC_P/mc^2)^{1/2}(\partial P/\partial e)_n = (\gamma - 1)^{1/2}$, where $\gamma = C_P/C_V$. We express the nonzero matrix elements $U_{ij}(\mathbf{k}, z)$ in terms of generalized transport coefficients, using (36) and (42).

For later convenience we also consider the case where i and/or j take the value l, although U_{il} and U_{li} are not elements of the matrix $U_{ij}(\mathbf{k}, z)$ of (36). We have now

$$U_{TT}(\mathbf{k}, z) = D_T(\mathbf{k}, z) = \lambda(\mathbf{k}, z)/nC_P \tag{44a}$$

$$U_{ll}(\mathbf{k}, z) = D_l(\mathbf{k}, z) \tag{44b}$$

$$U_{\eta_i\eta_i}(\mathbf{k}, z) = D_\eta(\mathbf{k}, z) = \eta(\mathbf{k}, z)/\rho \qquad (i = 1, 2) \tag{44c}$$

$$U_{lT}(\mathbf{k}, z) = U_{Tl}(\mathbf{k}, z) = (\rho c\alpha T)^{-1}(\gamma - 1)^{1/2}\theta(\mathbf{k}, z) \tag{44d}$$

$$U_{\sigma\sigma}(\mathbf{k}, z) = (1/2)(\gamma - 1)U_{TT}(\mathbf{k}, z) + (1/2)U_{ll}(\mathbf{k}, z) + \sigma(\gamma - 1)^{1/2}U_{Tl}(\mathbf{k}, z) \tag{44e}$$

$$U_{\sigma,-\sigma}(\mathbf{k}, z) = (1/2)(\gamma - 1)U_{TT}(\mathbf{k}, z) - (1/2)U_{ll}(\mathbf{k}, z) \tag{44f}$$

$$U_{T\sigma}(\mathbf{k}, z) = U_{\sigma T}(\mathbf{k}, z) = [(\gamma - 1)/2]^{1/2}U_{TT}(\mathbf{k}, z) + (\sigma/\sqrt{2})U_{Tl}(\mathbf{k}, z) \tag{44g}$$

where $\alpha = -n^{-1}(\partial n/\partial T)_P$ is the coefficient of thermal expansion, and in (44d) we used the thermodynamic identity $(\rho nTC_P)^{1/2} = \rho c\alpha T(\gamma - 1)^{-1/2}$. The generalized transport coefficients are defined as follows:

$$T\lambda(\mathbf{k}, z) = \beta(\hat{j}^e_{\mathbf{k}x}, \hat{\mathscr{G}}_z\hat{j}^e_{\mathbf{k}x}) \tag{45a}$$

$$\eta(\mathbf{k}, z) = \beta(\tau_{\mathbf{k}xy}, \hat{\mathscr{G}}_z\tau_{\mathbf{k}xy}) \tag{45b}$$

$$\rho D_l(\mathbf{k}, z) = \beta(\hat{\tau}_{\mathbf{k}xx}, \hat{\mathscr{G}}_z\hat{\tau}_{\mathbf{k}xx}) \tag{45c}$$

$$\theta(\mathbf{k}, z) = \beta(\hat{\tau}_{\mathbf{k}xx}, \hat{\mathscr{G}}_z\hat{j}^e_{\mathbf{k}x}) = \beta(\hat{j}^e_{\mathbf{k}x}, \hat{\mathscr{G}}_z\hat{\tau}_{\mathbf{k}xx}) \tag{45d}$$

Here $\lambda(\mathbf{k}, z)$ is the (generalized) coefficient of thermal conductivity, $\eta(\mathbf{k}, z)$ is the shear viscosity, $D_l(\mathbf{k}, z)$ is the longitudinal diffusivity,[3] and $\theta(\mathbf{k}, z)$ is a new transport coefficient describing the cross effects between the macroscopic heat current and the longitudinal momentum current [see Eqs. (137a) and (137b)].

We note in passing that $U_{ij}(\mathbf{k}, z)$ is not the Laplace transform of the time correlation function of two currents. Such a Laplace transform, denoted by $C_{ij}(\mathbf{k}, z)$, is given by

$$C_{ij}(\mathbf{k}, z) = (\hat{j}^i_{\mathbf{k}x}, \mathscr{G}_z\hat{j}^j_{\mathbf{k}x}) \tag{46}$$

and differs from $U_{ij}(\mathbf{k}, z)$ in that $C_{ij}(\mathbf{k}, z)$ contains the resolvent $\mathscr{G}_z = (z - L)^{-1}$ while $U_{ij}(\mathbf{k}, z)$ contains the projected resolvent $\hat{\mathscr{G}}_z$ given by (26).

[3] It does not seem meaningful to introduce, for $k \neq 0$, a k-dependent bulk viscosity, since the two obvious generalizations, either through the relation

$$\zeta(\mathbf{k}, z) = \rho D_l(\mathbf{k}, z) - \tfrac{4}{3}\eta(\mathbf{k}, z)$$

or through

$$\zeta'(\mathbf{k}, z) = \tfrac{1}{9}\beta \sum_{\alpha,\beta} (\hat{\tau}_{\mathbf{k}\alpha\alpha}, \hat{\mathscr{G}}_z\hat{\tau}_{\mathbf{k}\beta\beta}), \qquad \alpha, \beta = x, y, z$$

lead to different results which coincide only at $k = 0$.

Van Leeuwen and Ernst[10] and Zwanzig[8] have shown that the two matrices C_{ij} and U_{ij} are related to each other by the matrix relation

$$\mathbf{C} = (z\mathbf{1} + ik\mathbf{\Omega})\cdot(z\mathbf{1} + ik\mathbf{\Omega} + k^2\mathbf{U})^{-1}\cdot\mathbf{U} \tag{47}$$

which may be derived from the relations

$$\begin{aligned} \mathbf{U}(\mathbf{k}, z) &= \mathbf{C}(\mathbf{k}, z) + ik\mathbf{N}(\mathbf{k}, z)\cdot\mathbf{U}(\mathbf{k}, z) \\ z\mathbf{N}(\mathbf{k}, z) &= -ik\mathbf{C}(\mathbf{k}, z) - ik\mathbf{N}(\mathbf{k}, z)\cdot\mathbf{\Omega}(\mathbf{k}) \end{aligned} \tag{48}$$

where

$$N_{ij}(\mathbf{k}, z) = (\hat{j}^{i}_{\mathbf{k}x}, \mathscr{G}_z a_{\mathbf{k}}{}^{j}) \tag{49}$$

The component (η_i, η_i) of (47) has a particularly simple form, i.e.,

$$C_{\eta_i\eta_i}(\mathbf{k}, z) = zD_\eta(\mathbf{k}, z)[z + k^2 D_\eta(\mathbf{k}, z)]^{-1} \tag{50}$$

The connection between the Green–Kubo formulas for the Navier–Stokes transport coefficients and our formulas follows immediately from (47):

$$\lim_{z\to 0}\lim_{k\to 0} C_{ij}(\mathbf{k}, z) = \lim_{z\to 0}\lim_{k\to 0} U_{ij}(\mathbf{k}, z) \tag{51}$$

Consequently, in the limit indicated above, the generalized transport coefficients approach the usual time correlation function expressions for the Navier–Stokes transport coefficients λ, η, $\rho D_l = (4/3)\eta + \zeta$, where ζ is the bulk viscosity and $\lim \theta(\mathbf{k}, z) = 0$ as $k \to 0$.

So far we have obtained the Laplace transform of the hydrodynamic equations (30) and the equations (22) for the hydrodynamic propagators, together with explicit expressions for the quantities appearing in these equations. We are now interested in the behavior of the hydrodynamic variables $\langle n_{\mathbf{k}}(t)\rangle$, $\langle \mathbf{g}_{\mathbf{k}}(t)\rangle$, and $\langle e_{\mathbf{k}}(t)\rangle$ for small values of $\mathbf{k}$ and for large values of the time t. In order to find this behavior, we have to solve (30) and invert the Laplace transforms, i.e.,

$$\begin{aligned} \langle a_{\mathbf{k}}{}^{i}(t)\rangle = \oint_\Gamma \frac{dz}{2\pi i}\, e^{zt} \sum_j \{z\mathbf{1} + ik\mathbf{\Omega}(k) + k^2\mathbf{U}(\mathbf{k}, z)\}_{ij}^{-1} \\ \times \langle a_{\mathbf{k}}{}^{j}(0) + P_\perp \hat{\mathscr{G}}_z P_\perp L a_{\mathbf{k}}{}^{j}(0)\rangle \end{aligned} \tag{52}$$

The integral has to be taken along a path Γ in the complex z plane, running parallel to the imaginary axis and to the right of all singularities of the integrand in (52). In order to proceed, we have to investigate the analytic properties of $U_{ij}(\mathbf{k}, z)$ and $\langle P_\perp \hat{\mathscr{G}}_z P_\perp L a_{\mathbf{k}}{}^{j}(0)\rangle$ as functions of z around $z = 0$ for small values of the parameter k. If these functions were regular around $z = 0$, the long-time behavior of $a_{\mathbf{k}}{}^{i}(t)$ would be completely determined by the poles in the integrand (52) lying in the half-plane $\operatorname{Re} z < 0$, and lying closest to the imaginary axis (the hydrodynamic poles). The functions

$\langle a_{\mathbf{k}}^i(t)\rangle$ would behave as a sum of decaying exponentials, where the decay constants are given as the real parts of the hydrodynamic poles. The hydrodynamic poles are the roots of the secular determinant of the hydrodynamic matrix in (22) or (30), i.e.,

$$\det|z\,\delta_{ij} + ik\Omega_{ij}(\mathbf{k}) + k^2 U_{ij}(\mathbf{k}, z)| = 0 \tag{53}$$

which approach zero as k becomes small. These roots, $z_i(k)$, called the hydrodynamic frequencies, are found from (53) as functions $z_i(k)$ of k. The functions are called dispersion relations.

To lowest order in k, the hydrodynamic frequencies completely determine the long-time behavior of $\langle a_{\mathbf{k}}^i(t)\rangle$. This can be seen from (52), since $U_{ij}(\mathbf{k}, z)$ approaches the constant Navier–Stokes transport coefficients, and $\langle P_\perp \hat{\mathscr{G}}_z P_\perp L a_{\mathbf{k}}^j(0)\rangle = -ik\langle \Phi_N(0) P_\perp \hat{\mathscr{G}}_z \hat{j}_{\mathbf{k}x}^j\rangle_{\text{eq}}$ by virtue of (27) and (31). Thus this initial condition correction term is proportional to k, and can therefore be neglected with respect to $\langle a_{\mathbf{k}}^j(0)\rangle = \langle \Phi_N(0) a_{\mathbf{k}}^j\rangle_{\text{eq}}$ in (52).

If the functions $U_{ij}(\mathbf{k}, z)$ and $\langle \Phi_N(0) P_\perp \hat{\mathscr{G}}_z \hat{j}_{\mathbf{k}x}^j\rangle_{\text{eq}}$ are not regular around $z = 0$, one needs to know the locations and nature of their singularities, such as branch points. This subject will be discussed in Section 4, while Section 3 is devoted to the dispersion relations. Henceforth we will neglect the initial condition correction term in (52), since it is proportional to k in the limit of small z and k, and its coefficient $\langle \Phi_N(0) P_\perp \hat{\mathscr{G}}_0 \hat{j}_{0x}^j\rangle_{\text{eq}}$ is simply a constant. This coefficient has basically the same structure as the matrix elements $U_{ij}(\mathbf{k}, z)$, which are known to exist for $k = z = 0$ in three-dimensional fluids, and for consistency with other approximations, any correction terms proportional to k should be neglected. In Section 4 it will also be shown that the hydrodynamic frequencies determine the long-time behavior of $\langle a_{\mathbf{k}}^i(t)\rangle$ to the order of k, in which we are interested.

3. DISPERSION RELATIONS

In this section we will determine the hydrodynamic frequencies for small values of k by solving the secular equation (53). To do so, we will use the phenomenological mode–mode coupling theory to describe the behavior of $U_{ij}(\mathbf{k}, z)$ for small values of k and z. In Ref. 1 we showed that there exist nonanalytic terms in the k expansion of the hydrodynamic frequencies for a gas, and later we extended[11] these results to fluids. The general form of the dispersion relations obtained was

$$z_i(k) = ak + bk^2 + ck^{5/2} + \cdots \tag{54}$$

This result was also obtained by Pomeau,[6] who showed that there is an infinite number of terms between $k^{5/2}$ and k^3 which have the general form k^{2+P_n}, where $P_n = 1 - 2^{-n}$, $n = 1, 2, \ldots$. Here we give in detail the method that leads to (54) for a general fluid, and also obtain the terms of the general form as found by Pomeau, but with a different set of coefficients. In the final section we will present a comparison of our results with Pomeau's, and in Section 5 we present an analysis of the convergence of the series.

First, we consider the secular determinant (53). Since there are no matrix elements $U_{ij}(\mathbf{k}, z)$ connecting the labels η_1 and η_2 with one another or with T and σ, this determinant factors into a (3×3) determinant with labels $(i, j) = (T, \sigma = \pm)$ and two identical equations labeled with η_1 and η_2, namely

$$z + k^2 U_{\eta_i\eta_i}(\mathbf{k}, z) = z + k^2 D_\eta(\mathbf{k}, z) = 0 \tag{55}$$

The solution $z_\eta(k)$ which approaches zero as $k \to 0$ is the shear mode frequency or relaxation rate of two degenerate shear modes $a_{\mathbf{k}}^{\eta_1}$ and $a_{\mathbf{k}}^{\eta_2}$, which are given to lowest order in k by (17c).

The frequencies of the three remaining hydrodynamic modes are obtained by solving the (3×3) secular determinant, where $(i, j) = (T, \sigma = \pm)$. Since the off-diagonal elements are at least of order k^2 for small z, the eigenvalues are determined, up to terms of $O(k^3)$, by solving the following equations:

$$z + k^2 U_{TT}(\mathbf{k}, z) = z + k^2 D_T(\mathbf{k}, z) = 0 \tag{56}$$

and the two sound mode frequencies are determined by the two equations $(\sigma = \pm)$

$$z + ik\Omega_{\sigma\sigma}(\mathbf{k}) + k^2 U_{\sigma\sigma}(\mathbf{k}, z) = z + i\sigma ck + k^2 U_{\sigma\sigma}(\mathbf{k}, z) = 0 \tag{57}$$

To lowest order in k the corresponding heat mode and sound modes are given by (17). Of course, the hydrodynamic frequencies depend neither on the normalization (16) nor on the particular linear combinations of $n_{\mathbf{k}}$, $g_{\mathbf{k}}$, and $e_{\mathbf{k}}$ used here. In fact, in Section 6 we will discuss different linear combinations of these variables in connection with the Burnett and super-Burnett equations. The corresponding secular determinant reduces again to (55)–(57), since the corresponding matrices are related by a similarity transformation.

We also want to point out that the generalized sound wave damping constant $U_{\sigma\sigma}(\mathbf{k}, z)$ differs from the usual expression $D_s = \frac{1}{2}(\gamma - 1)D_T + \frac{1}{2}D_l$ by the occurrence of the new transport coefficient $\theta(\mathbf{k}, z)$, given in (45d). As we noted already, $\theta(\mathbf{k}, z)$ vanishes as $k \to 0$, since in this limit it is an average of an odd function of the velocities. If $\theta(\mathbf{k}, z)$ were regular in k and z around the origin, it could be expanded in a Taylor series around $z = k = 0$, and the first nonvanishing contribution would give rise to a term of $O(k^3)$ in $z_\sigma(k)$. If this were the case, the term $\theta(\mathbf{k}, z)$ should be neglected in (44e) for consistency. We will show, however, that $\theta(\mathbf{k}, z)$ contains contributions of $O(k^{1/2})$, and should therefore be kept in (44e).

The general form of the equations to be solved is now

$$z + ik\Omega_{ii} + k^2 U_{ii}(\mathbf{k}, z) = 0 \tag{58}$$

or more specifically

$$z + k^2 D_\mu(\mathbf{k}, z) = 0, \qquad \mu = \eta, T \tag{58a}$$

$$z + i\sigma ck + k^2 U_{\sigma\sigma}(\mathbf{k}, z) = 0, \qquad \sigma = \pm \tag{58b}$$

We first observe that complex conjugation of these equations leads to

$$\begin{aligned} z^* + k^2 D_\mu(\mathbf{k}, z^*) &= 0 \\ z^* - i\sigma ck + k^2 U_{-\sigma,-\sigma}(\mathbf{k}, z^*) &= 0 \end{aligned} \tag{59}$$

This can be seen by observing that D_μ and D_l in (45) satisfy $D^*(\mathbf{k}, z) = D(\mathbf{k}, z^*)$, while $\theta^*(\mathbf{k}, z) = -\theta(\mathbf{k}, z^*)$.

Equation (58) will be solved by successive approximation. The solution of (58) to lowest order in k can be found directly by replacing $U_{ii}(\mathbf{k}, z)$ by $\lim_{k\to 0} U_{ii}(\mathbf{k}, z)$ and then solving (58) for z to lowest (called zeroth) order in k. The result is

$$z_i^0(k) = -ik\Omega_{ii} - k^2 U_{ii} \tag{60}$$

or more specifically

$$z_\mu^0 = -k^2 D_\mu \tag{60a}$$

$$z_\sigma^0 = -i\sigma ck - k^2 D_s \tag{60b}$$

where

$$U_{ij} = \lim_{z\to 0} \lim_{k\to 0} U_{ij}(\mathbf{k}, z) \tag{61}$$

and similar definitions for D_μ ($\mu = \eta, T$) and D_s. Equations (60a) and (60b) are the hydrodynamic frequencies obtained from the usual Navier–Stokes equations, as follows from (45), (46), and (51). We now assume that the solutions of (58a) and (58b) of interest to us, i.e., those that reduce to the values given by Eqs. (60a) and (60b), respectively, for small k, are unique. This assumption, together with Eq. (59), leads to the result that the solution $z_\mu(k)$ of (58a) is real, while the solution $z_\sigma(k)$ of Eq. (58b) satisfies $z_\sigma^* = z_{-\sigma}$.

To determine the next approximation to $z_i(k)$, we have to know the behavior of $U_{ii}(\mathbf{k}, z)$ for small values of k and z. Consider, therefore, the Taylor series expansion

$$\begin{aligned} U_{ii}(\mathbf{k}, z) = U_{ii}(0, 0) &+ k\left(\frac{\partial U_{ii}}{\partial k}\right)_{k\to 0, z\to 0} + z\left(\frac{\partial U_{ii}}{\partial z}\right)_{k\to 0, z\to 0} \\ &+ \frac{1}{2}\left(k^2 \frac{\partial^2 U_{ii}}{\partial k^2} + 2kz \frac{\partial^2 U_{ii}}{\partial k\, \partial z} + z^2 \frac{\partial^2 U_{ii}}{\partial z^2}\right)_{k\to 0, z\to 0} + \cdots \end{aligned} \tag{62}$$

However, the double expansion does not exist. This is a consequence of the long-time behavior of the current–current correlation functions $\tilde{C}_{ii}(0, t)$, which are the inverse Laplace transforms of $C_{ii}(0, z)$ defined in (46). It was found[1,5] that $\tilde{C}_{ii}(0, t)$ behaves asymptotically as $t^{-3/2}$, which implies that for small z the functions $C_{ii}(0, z)$—and by virtue of (47) also $U_{ii}(0, z)$—behave as

$$C_{ii}(0, z) = U_{ii}(0, z) \simeq C_{ii}(0, 0) + a\sqrt{z} \tag{63}$$

where a is some constant. Consequently, $\partial U_{ii}/\partial z$ for $k = z = 0$ does not exist, and neither does the expansion (62). The function $U_{ii}(\mathbf{k}, z)$ is therefore singular around $k = z = 0$. Let us investigate the implications of these singularities in $k = z = 0$ for the solutions of (58). To find the first correction to the lowest order solution (60), one would be inclined to insert (63) into (58), and conclude that $z_\mu(k) = -k^2 D_\mu + ak^3$ and $z_\sigma = -i\sigma ck - k^2 D_s + bk^{5/2}$, where a and b are some constants. However, this is not correct for $z_\mu(k)$, and although it is qualitatively correct for $z_\sigma(k)$, it does not lead to the correct coefficient. This is because to solve (58a) and (58b), one needs the behavior of $U_{ii}(k, z)$ as a function of both k and z in regions where $z \sim k^2$ or $z \sim k$ and k is small, but one does not need the behavior of $U_{ii}(0, z)$ for small values of z, as given in (63).

We now make the *basic assumption* that the leading singularities of $U_{ij}(\mathbf{k}, z)$ in the small (k, z) regions of interest are given by the *mode–mode coupling theory*. This theory leads to explicit expressions for $U_{ij}(\mathbf{k}, z)$ in the (k, z) regions of interest, and we will show that they lead to a non-power series expansion of the hydrodynamic frequencies.[4] The mode–mode coupling theory yields the following expression for the part of $U_{ij}(\mathbf{k}, z)$ that contains the dominant singularities in k and z:

$$U_{ij}(\mathbf{k}, z) = U_{ij}^0 + \tfrac{1}{2} \sum_{a,b} U_{ij}^{ab}(\mathbf{k}, z) \tag{64a}$$

$$= U_{ij} + \tfrac{1}{2} \sum_{a,b} \delta U_{ij}^{ab}(\mathbf{k}, z) \tag{64b}$$

The sum runs over all pairs of hydrodynamic modes

$$(a, b) = (T, \eta_1, \eta_2, \sigma = \pm);$$

U_{ij}^0 is the so called *bare* transport coefficient; and $U_{ij}^{ab}(\mathbf{k}, z)$ are the mode–mode integrals

$$U_{ij}^{ab}(\mathbf{k}, z) = \int' \frac{d\mathbf{q}}{(2\pi)^3} \frac{S_{ij}^{ab}(\hat{\mathbf{q}}, \hat{\mathbf{l}})}{z - z_a(q) - z_b(l)} \tag{65}$$

[4] It should be pointed out that the mode–mode formula applies only to the functions $U_{ij}(\mathbf{k}, z)$, which contain the projected operator $\hat{\mathscr{G}}_z$, and not to the correlation functions $C_{ij}(\mathbf{k}, z)$, where the unprojected operator $\mathscr{G}_z$ appears. For a discussion of this point see Ernst *et al.*[12]

The prime on the integral sign indicates that $|\mathbf{q}| < k_0$, where k_0 is a cutoff wave number of the order of a reciprocal microscopic correlation length—the inverse mean free path in a dilute gas, and the inverse range of the intermolecular potential in a fluid. Furthermore, $\mathbf{l} = \mathbf{k} - \mathbf{q}$; $\hat{\mathbf{q}}$ and $\hat{\mathbf{l}}$ are unit vectors; and $\mathbf{k}$ is always taken parallel to the x axis. The strength factor $S_{ij}^{ab}(\hat{\mathbf{q}}, \hat{\mathbf{l}})$ is given in terms of the mode–mode amplitude $A_i^{ab}(\hat{\mathbf{q}}, \hat{\mathbf{l}})$ as

$$S_{ij}^{ab}(\hat{\mathbf{q}}, \hat{\mathbf{l}}) = A_i^{ab}(\hat{\mathbf{q}}, \hat{\mathbf{l}})(A_j^{ab}(\hat{\mathbf{q}}, \hat{\mathbf{l}}))^* \tag{66}$$

with

$$A_i^{ab}(\hat{\mathbf{q}}, \hat{\mathbf{l}}) = (\hat{j}_{\mathbf{k}x}^{i}, a_{\mathbf{q}}{}^{a} a_{\mathbf{l}}{}^{b}) \tag{67}$$

where $a_{\mathbf{k}}{}^{b}$ are the hydrodynamic modes given in (17). The bare transport coefficient U_{ij}^{0} in (64a) may be eliminated from (64a) by virtue of (61), where we identify the Navier–Stokes transport coefficient U_{ij} with

$$U_{ij} = U_{ij}^{0} + \tfrac{1}{2} \sum_{a,b} U_{ij}^{ab}(0, 0) \tag{68}$$

We have used the fact that $U_{ij}^{ab}(0, 0)$ in (65) does not depend on the order in which k and z approach zero. Therefore (64a) can be written in the alternative form (64b), which will be used in this paper; and $\delta U_{ij}^{ab}(\mathbf{k}, z)$ is defined as

$$\delta U_{ij}^{ab}(\mathbf{k}, z) = U_{ij}^{ab}(\mathbf{k}, z) - U_{ij}^{ab}(0, 0) \tag{69}$$

The mode–mode formula is well known and frequently used in analyzing the singular behavior of transport coefficients near critical points. Derivations of this formula based on intuitive arguments have been given by Kadanoff and Swift,[2] Kawasaki,[3] and Ferrell.[4] Phenomenological arguments for its validity away from the critical point have also been published.[5] A more rigorous approach, valid for hard sphere gases, was given in Ref. 1.

On the basis of the above assumption (64a)–(64b), the dispersion relations (58) for small values of k can be written as

$$z_i(k) = -ik\Omega_{ii} - k^2 U_{ii} - \tfrac{1}{2} \sum_{a,b} k^2\, \delta U_{ii}^{ab}(\mathbf{k}, z_i(k)) \tag{70}$$

This equation is in fact a complicated set of coupled nonlinear integral equations for $z_i(k)$ with $i = \sigma, \eta, T$, where all unknown frequencies enter again in the denominators of the mode–mode integrals. We have not been able to find a solution of these equations in closed form, but we present a scheme for successive approximations, valid for small values of k.

In Appendix B we analyze the mode–mode integrals $U_{ij}^{ab}(\mathbf{k}, z)$ and we find that the dominant contributions come either from two oppositely traveling sound modes, i.e., $U_{ij}^{\sigma-\sigma}(\mathbf{k}, z)$ with $\sigma = \pm$, or from two diffusive modes, i.e., $\delta U_{ij}^{\lambda\mu}(\mathbf{k}, z)$ with $(\lambda, \mu) = (T, \eta_1, \eta_2)$. The first approximation to the solution of (70) is now obtained by replacing all hydrodynamic frequencies in the denominator of (65) by their zeroth approximation $z_a{}^0$ given in (60). In addition, we replace the argument $z_i(k)$ of δU_{ij}^{ab} by $z_i{}^0(k)$. From Eqs. (B.2), (B.8), and (B.10)–(B.14) of Appendix B we have now in zeroth approximation

$$\delta U_{ii}^{\sigma-\sigma}(\mathbf{k}, z_i{}^0) = -\int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S_{ii}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{4\pi(2D_s)^{3/2}} (z_i{}^0 + i\sigma ck\hat{q}_x)^{1/2} + O(k) \quad (71a)$$

$$\delta U_{ii}^{\lambda\mu}(\mathbf{k}, z_i{}^0) = -\int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S_{ii}^{\lambda\mu}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{4\pi(2D_{\lambda\mu})^{3/2}} (z_i{}^0)^{1/2} + O(k) \quad (71b)$$

Here terms of $O(k)$ have been neglected, $d\hat{\mathbf{q}}$ is a solid angle, with $\hat{q}_x = \hat{\mathbf{q}}\cdot\hat{\mathbf{k}}$. We have further introduced

$$D_{\lambda\mu} = \tfrac{1}{2}(D_\lambda + D_\mu) \quad (72)$$

Consider first (70) for the diffusive modes ($\mu = \eta, T$), where $z_\mu{}^0 = -k^2 D_\mu$. In this case the term $z_i{}^0$ in (71a) should be neglected compared to $i\sigma ckq_x$ for consistency, whereas the contributions from two diffusive modes (71b) should be neglected completely, since the first term in (71b) is as unimportant as the neglected terms of $O(k)$. We then find from (70) and (71) the hydrodynamic frequency $z_\mu{}^1$ of the diffusive modes in first approximation,

$$z_\mu{}^1 = -k^2 D_\mu + \frac{1}{2} k^2 \sum_\sigma \int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S_{\mu\mu}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{4\pi(2D_s)^{3/2}} (i\sigma ck\hat{q}_x)^{1/2} \quad (73)$$

From the explicit expressions for the function $S(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$, calculated in (C.4) and (C.6) of Appendix C, one sees that $S_{\mu\mu}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ is a real, even function of $\hat{q}_x$, so that we can replace $(i\sigma\hat{q}_x)^{1/2}$ in (73) by $2^{-1/2}|\hat{q}_x|^{1/2}$. This yields the first approximation to the frequency of the diffusive modes

$$z_\mu{}^1 = -k^2 D_\mu + k^{5/2}\Delta_\mu(1) \quad (74)$$

with

$$\Delta_\mu(1) = \frac{1}{4\pi\sqrt{2}} \frac{\sqrt{c}}{\beta\rho} \frac{M_{\mu\mu}^{ss}(1)}{(2D_s)^{3/2}} \quad (75)$$

and we have introduced for later convenience the quantity $M_{\mu\mu}^{ss}(n)$, where $n = 1$ in (75), as

$$M_{\mu\mu}^{ss}(n) = \frac{1}{2}\beta\rho \sum_\sigma \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{\mu\mu}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})|\hat{q}_x|^{P_n} \quad (76)$$

with

$$P_n = 1 - 2^{-n} \quad (77)$$

so that $P_1 = 1/2$. The superscripts (ss) refer to the sound modes. The integrals in (76) are carried out in Eqs. (C.20)–(C.21) of Appendix C.

Consider now the frequency of the sound mode in (70). In this case z_i^0 in the mode–mode integrals (71) should be taken as $-i\sigma ck$ by virtue of (60b), and we obtain

$$z_\sigma{}^1 = -i\sigma ck - k^2 D_s + \frac{1}{2}k^2 \sum_{\lambda\mu} \int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S_{\sigma\sigma}^{\lambda\mu}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{4\pi(2D_{\lambda\mu})^{3/2}} (-i\sigma ck)^{1/2}$$

$$+ \frac{1}{2}k^2 \sum_{\sigma'} \int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S_{\sigma\sigma}^{\sigma'-\sigma'}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{4\pi(2D_s)^{3/2}} (1 - \sigma\sigma'\hat{q}_x)^{1/2}(-i\sigma ck)^{1/2} \tag{78}$$

Using Eqs. (C.11)–(C.14) of Appendix C, one sees that $S_{\sigma\sigma}^{\lambda\mu}$ does not depend on σ, and that

$$S_{\sigma\sigma}^{\sigma'-\sigma'}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}M(\sigma\sigma'\hat{q}_x) \tag{79}$$

where $M(x)$, given in (C.14), is a real, positive function of x. This gives us the first approximation for the sound mode frequency

$$z_\sigma{}^1 = -i\sigma ck - k^2 D_s + k^{5/2}[\Delta_s(1) - i\sigma\bar{\Delta}_s(1)] \tag{80}$$

Here

$$\Delta_s(1) = \bar{\Delta}_s(1) = \frac{1}{4\pi\sqrt{2}} \frac{\sqrt{c}}{\beta\rho} \sum_{\langle ab\rangle} \frac{M_{\sigma\sigma}^{ab}(1)}{(2D_{ab})^{3/2}} \tag{81}$$

where the sum runs over the four terms $\langle ab\rangle = (ss)$, (η, η), (ηT), and (TT). Again we introduce for later convenience the quantities $M_{\sigma\sigma}^{ab}(n)$, where $n = 1$ in (81), as

$$M_{\sigma\sigma}^{\eta\eta}(n) = M_{\sigma\sigma}^{\eta\eta}(0) = \frac{1}{2}\beta\rho \sum_{i,j=1,2} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{\sigma\sigma}^{\eta_i\eta_j}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \tag{82}$$

$$M_{\sigma\sigma}^{\eta T}(n) = M_{\sigma\sigma}^{\eta T}(0) = \beta\rho \sum_{i=1,2} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{\sigma\sigma}^{\eta_i T}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \tag{83}$$

$$M_{\sigma\sigma}^{TT}(n) = M_{\sigma\sigma}^{TT}(0) = \frac{1}{2}\beta\rho \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{\sigma\sigma}^{TT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \tag{84}$$

$$M_{\sigma\sigma}^{ss}(n) = \frac{1}{2}\beta\rho \sum_{\sigma'} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{\sigma\sigma}^{\sigma'-\sigma'}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})(1 - \sigma\sigma'\hat{q}_x)^{p_n} \tag{85a}$$

$$= \frac{1}{2}\int_{-1}^{1} dx\, M(x)(1 - x)^{p_n} \tag{85b}$$

In (85a)–(85b) we have used (79). The functions $S_{\sigma\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ are calculated in Appendix C, where the angular integrations in (82)–(85) are carried out. At this point one can verify that $\theta(\mathbf{k}, z)$, defined by Eq. (45d), is proportional to $k^{1/2}$. This can be seen by applying the mode–mode coupling formula to (45d) and considering the contribution from antiparallel sound modes, using the method outlined in Appendix B for treating the mode–mode integrals.

This procedure leads directly to the result that $\theta(\mathbf{k}, z) \sim k^{1/2}$ for small k, and in fact shows that $\theta(\mathbf{k}, z)$ forms part of the contribution to the coefficient of $k^{5/2}$ in Eq. (80).

So far we have obtained the first approximation (74) and (80) for the hydrodynamic frequencies, and explicit expressions for the coefficients are given in (75), (81), (C.20), (C.21), (C.24), and Table I of Appendix C. These results were given in a previous publication,[11] where a printing error occurs in the expression for $M^{ss}_{\sigma\sigma}(1)$ (indicated there by M_{ss}); this has been corrected in (C.24).

In the next part of this section we discuss the solutions of (70) obtained by successive higher approximations. The approximation scheme can be simply formulated with the results of Appendix B. In this appendix we have made the following ansatz for the hydrodynamic frequencies appearing in (65):

$$\begin{aligned} z_\mu(k) &= -k^2 D_\mu + \Delta_\mu k^{2+P} \\ z_\sigma(k) &= -i\sigma ck - k^2 D_s + (\Delta_s - i\sigma\bar{\Delta}_s)k^{2+P} \end{aligned} \tag{86}$$

and we calculated the mode–mode integral (65) with the result

$$\delta U^{\sigma-\sigma}(\mathbf{k}, z_i(k)) = -\int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{8\pi D_s} \left\{ \zeta_1^{1/2} + \frac{\Delta_s}{D_s} \frac{P+3}{2\cos\frac{1}{2}\pi P} \zeta_1^{(P+1)/2} \right\} + O(k) \tag{87a}$$

$$\delta U^{\lambda\mu}(\mathbf{k}, \mathbf{z}_i(k)) = -\int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S^{\lambda\mu}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{8\pi D_{\lambda\mu}} \left\{ \zeta_2^{1/2} + \frac{\Delta_{\lambda\mu}}{D_{\lambda\mu}} \frac{P+3}{2\cos\frac{1}{2}\pi P} \zeta_2^{(P+1)/2} \right\} + O(k) \tag{87b}$$

where $\Delta_{\lambda\mu} = \frac{1}{2}(\Delta_\lambda + \Delta_\mu)$ and

$$\zeta_1 = [z_i(k) + i\sigma ck\hat{q}_x]/2D_s, \qquad \zeta_2 = z_i(k)/2D_{\lambda\mu} \tag{88}$$

We consider first (70) for the sound mode frequency where $z_i(k) = z_\sigma(k)$. Here we see that the dispersion relations (86), used as input to calculate $\delta U^{\sigma'-\sigma'}(\mathbf{k}, z(k))$ and $\delta U^{\lambda\mu}(\mathbf{k}, z_\sigma(k))$, produce in (87) a term of $O(k^{1/2})$ and $O(k^{(P+1)/2})$. Exactly the same thing happens for the frequencies of the diffusive modes $z_i(k) = z_\mu(k)$ with $\mu = \eta$, T, where only $\delta U^{\sigma-\sigma}(\mathbf{k}, z_\sigma(k))$ has to be considered for solving (70), since $\delta U^{\lambda\mu}(\mathbf{k}, z_\mu(k))$ is of $O(k)$.

Therefore, if we had taken $P = \frac{1}{2}$ in (86) we would have found terms of $O(k^{1/2})$ and $O(k^{3/4})$ in (87). If we had used this result again as input, we would have obtained from (87) terms of $O(k^{1/2})$, $O(k^{3/4})$, and $O(k^{7/8})$. The general structure is therefore that a term of $O(k^{P_n})$ with $P_n = 1 - 2^{-n}$ in the dispersion relation (86) produces through iteration of the mode–mode formula a new term of $O(k^{(P_n+1)/2}) = O(k^{P_{n+1}})$. In order to make the whole scheme self-consistent we write the dispersion relations as

$$z_\mu(k) = -D_\mu k^2 + \sum_{n=1} \Delta_\mu(n) k^{2+P_n} \tag{89a}$$

$$z_\sigma(k) = -i\sigma ck - D_s k^2 + \sum_{n=1} [\Delta_s(n) - i\sigma\bar{\Delta}_s(n)] k^{2+P_n} \tag{89b}$$

with $P_n = 1 - 2^{-n}$. The approximation scheme outlined above will automatically lead to recursion relations between the coefficients $\Delta_i(n)$, which we derive now. We calculate $z_i(k)$ from (70) by means of the mode–mode integrals (87), in which we use (89) as input. The analysis of the mode–mode integrals of Appendix B still applies, since the exponents P_n satisfy the requirement $\frac{1}{2} \leqslant P_n < 1$ (which is necessary in Appendix B), and we replace Δ and P in (86) by $\Delta(n)$ and P_n, respectively, and sum over $n = 1, 2, \ldots$. For the diffusive modes ($\mu = \eta, T$) we obtain from (87)–(89) in this way

$$z_\mu(k) = -D_\mu k^2 + \frac{1}{2} k^2 \sum_\sigma \int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S_{\mu\mu}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{4\pi D_s^2}$$

$$\times \sum_{n=0} \Delta_s(n) \frac{P_{n+2}}{\cos\frac{1}{2}\pi P_n} \left(\frac{i\sigma ck\hat{q}_x}{2D_s}\right)^{P_{n+1}} \tag{90}$$

In the summation over n we have included the term obtained in (73) by putting $n = 0$ and defining

$$\Delta_a(0) = \tfrac{2}{3} D_a, \qquad a = s, \eta, T \tag{91}$$

From the relation $i^{P_n} = \cos\frac{1}{2}\pi P_n + i \sin\frac{1}{2}\pi P_n$, and from arguments similar to those used following (73), we deduce

$$z_\mu = -D_\mu k^2 + \sum_{n=0} \Delta_s(n) L_\mu^{ss}(n) \frac{\cos\frac{1}{2}\pi P_{n+1}}{\cos\frac{1}{2}\pi P_n} k^{2+P_{n+1}} \tag{92}$$

with

$$L_\mu^{ss}(n) = \frac{P_{n+2}}{4\pi\beta\rho D_s^2} \left(\frac{c}{2D_s}\right)^{P_{n+1}} M_{\mu\mu}^{ss}(n+1) \tag{93}$$

where $M_{\mu\mu}^{ss}(n)$ is defined in (76) and calculated in (C.19)–(C.21). For the sound mode frequency we obtain in a similar way

$$z_\sigma = -i\sigma ck - D_s k^2 + k^2 \sum_{\langle ab\rangle} \sum_{n=0} \Delta_{ab}(n) L_s^{ab}(n) \frac{(-i\sigma k)^{P_{n+1}}}{\cos\frac{1}{2}\pi P_n} \tag{94}$$

where $\langle ab\rangle = (ss), (\eta\eta), (\eta T)$, and (TT), and

$$\Delta_{ab}(n) = \tfrac{1}{2}[\Delta_a(n) + \Delta_b(n)], \qquad D_{ab} = \tfrac{1}{2}(D_a + D_b) \tag{95}$$

with $a = (s, \eta, T)$. Furthermore,

$$L_s^{ab}(n) = \frac{P_{n+2}}{4\pi\beta\rho D_{ab}^2} \left(\frac{c}{2D_{ab}}\right)^{P_{n+1}} M_{\sigma\sigma}^{ab}(n+1) \tag{96}$$

where $M_{\sigma\sigma}^{ss}(n)$ and $M_{\sigma\sigma}^{\lambda\mu}(n)$ are defined in (82)–(85) and calculated in Appendix C, starting from (C.21) on.

By comparing the expressions for $z_i(k)$ in (89), (92), and (94), and equating (the real and imaginary parts of) the coefficients of equal powers of k, one obtains the recursion relations for $n \geqslant 0$:

$$\delta_\mu(n+1) = L_\mu^{ss}(n)\,\delta_s(n) \tag{97a}$$

$$\delta_s(n+1) = \sum_{\langle ab\rangle} L_s^{ab}(n)\,\delta_{ab}(n) \tag{97b}$$

where we have introduced

$$\begin{aligned} \Delta_a(n) &= \delta_a(n)\cos\tfrac{1}{2}\pi P_n, \qquad a = s, \eta, T \\ \bar{\Delta}_s(n) &= \delta_s(n)\sin\tfrac{1}{2}\pi P_n = \Delta_s(n)\tan\tfrac{1}{2}\pi P_n \end{aligned} \tag{98}$$

and

$$\delta_{ab}(n) = \tfrac{1}{2}[\delta_a(n) + \delta_b(n)] \tag{99}$$

and $\delta_a(0) = \frac{2}{3}D_a$, as follows from (98) and (91).

These recursion relations can be solved successively. It follows directly that all $\Delta_a(n)$ are positive, since $L(n)$ is positive. For $n = 0$, Eq. (97) yields (74) and (80), which we already obtained by explicit calculation. Using the expressions for the coefficients $M(n)$ calculated in Appendix C, we can directly obtain explicit expressions for $L(n)$ and $\Delta_a(n)$. It is also interesting to observe that $z_\sigma(k) \equiv \omega_s(-i\sigma k)$ is related to a real function $\omega_s(k)$, defined as

$$\omega_s(x) = cx + D_s x^2 - \sum_{n=1} \delta_s(n) x^{2+P_n} \tag{100}$$

and similarly $z_\mu(k) = \operatorname{Re}\omega_\mu(ik)$, where

$$\omega_\mu(x) = D_\mu x^2 - \sum_{n=1} \delta_\mu(n) x^{2+P_n} \tag{101}$$

The general structure of the recursion relations (97) was obtained by Pomeau.[6] We disagree, however, with his results for the coefficients $L(n)$ for several reasons, to be discussed in the last section.

4. BRANCH POINTS OF $U_{ij}(\mathbf{k},z)$

In the previous section we discussed the contribution to $\langle a_{\mathbf{k}}^i(t)\rangle$ from poles in the integrand on the right-hand side of Eq. (52), i.e., from zeros of the hydrodynamic matrix. In addition to the poles, the integrand has branch point singularities which must be taken into account when inverting the Laplace transform.[7] To see that there are branch points in $U_{ij}(\mathbf{k}, z)$, consider Eq. (B.10) of Appendix B. Inspection of $U^{\sigma-\sigma}(\mathbf{k}, z)$ given by this equation shows that there are branch points in the complex z plane at the values

$z = \pm ick$, and the presence of these branch points requires that cuts be made in the z plane along the lines $-\infty \leqslant z + ick \leqslant 0$. Since the long-time behavior of $\langle a_{\mathbf{k}}{}^{i}(t)\rangle$ may also be determined by the branch point singularities, we will consider whether the contributions from the branch points are of the same order of magnitude as those coming from the poles.

In the previous section we found that there are poles in the hydrodynamic matrix in two regions in the z plane, (a) $z \approx \pm ikc$ and (b) $z \approx -Dk^2$, where D is an appropriate diffusivity. Connected to these poles there are two time scales, (a) $\tau_s(k)$, the period of oscillation of a sound wave of wave number k, where $\tau_s(k) \simeq (ck)^{-1}$, and (b) $\tau_d(k)$, the relaxation time for the decay of a sound, shear, or thermal mode of wave number k, where $\tau_d(k) \simeq (Dk^2)^{-1}$. Since $\tau_s(k) \ll \tau_d(k)$ for small k, there is practically no damping of a sound, shear, or thermal mode on the time scale of $\tau_s(k)$. To estimate the relative sizes of the contributions to $\langle a_{\mathbf{k}}{}^{i}(t)\rangle$ from the poles and the branch point singularities, we must determine (a) whether there are any branch points in $U(\mathbf{k}, z)$ in the same regions of the z plane as hydrodynamic poles, (b) the relative order of the contributions to $\langle a_{\mathbf{k}}{}^{i}(t)\rangle$ from each type of singularity, if there are any, and (c) whether any contribution from branch points are more important than those from poles. Therefore we will consider the analytic properties of $U(\mathbf{k}, z)$ as a function of z for fixed but small k.

First we consider question (a), and we will be interested then in locating any branch points in $U(\mathbf{k}, z)$ that may lie in one of three regions of the z plane

$$\begin{aligned} &\text{(i)} \quad z \sim O(k) \\ &\text{(ii)} \quad z \sim O(k^2) \\ &\text{(iii)} \quad z \ll O(k^2) \end{aligned} \tag{102}$$

Region (i) will determine branch point contributions to $\langle a_{\mathbf{k}}{}^{i}(t)\rangle$ on the time scale of a sound oscillation $\tau_s(k)$; region (ii), contributions on the time scale of a damping time $\tau_d(k)$; and region (iii), contributions on any scale which is longer than $\tau_d(k)$.

We begin by noting that there are four distinct types of contributions to $U_{ij}(\mathbf{k}, z)$ coming from combinations of two hydrodynamic modes. We have denoted these by $\delta U_{ij}^{\sigma,-\sigma}(\mathbf{k}, z)$, $\delta U_{ij}^{\lambda\mu}(\mathbf{k}, z)$, $\delta U_{ij}^{\sigma\lambda}(\mathbf{k}, z)$, and $\delta U_{ij}^{\sigma\sigma}(\mathbf{k}, z)$, coming from antiparallel sound modes, two diffusive modes, a diffusive and a sound mode, and parallel sound modes, respectively. We consider first the quantity $\delta U_{ij}^{\sigma,-\sigma}$, which is given, for small k and z, by Eq. (B.10) of Appendix B, with the dispersion relations (89),

$$\delta U_{ij}^{\sigma,-\sigma}(\mathbf{k}, z) = -\frac{1}{4\pi D_s}\int \frac{d\hat{\mathbf{q}}}{4\pi}\, S_{ij}^{\sigma,-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})\Bigg\{\zeta^{1/2} + \sum_{n=1} \frac{\Delta_s(n)}{D_s}\,\frac{P_{n+2}}{\cos\frac{1}{2}\pi P_n}\,\zeta^{P_{n+1}}\Bigg\} + O(\theta) \tag{103}$$

Here $\theta = \max(z, k)$, $P_n = 1 - 2^{-n}$, and $\zeta = (z + i\sigma ckx)/2D_s$, where $x = \hat{k}\cdot\hat{q} = \hat{q}_x$. Using the fact that $S(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ is only a polynomial in x [see Eqs. (C.4)–(C.17)], we conclude that $\delta U_{ij}^{\sigma-\sigma}(\mathbf{k}, z)$ contains terms proportional to $(z \pm i\sigma ck)^{1/2}$ and $(z \pm i\sigma ck)^{P_{n+1}}$, each multiplied by a polynomial in (z/k). Consequently, $\delta U_{ij}^{\sigma-\sigma}(\mathbf{k}, z)$ has branch points in the complex z plane at $z = \pm ick$. Having located branch points in $\delta U^{\sigma-\sigma}(\mathbf{k}, z)$ for $z \simeq O(k)$, we expect that these branch points will contribute to $\langle a_{\mathbf{k}}{}^i(t)\rangle$ for times of the order of $\tau_s(k)$. The correction term of $O(\theta)$ in (103) is of $O(k)$, since on the time scales of interest $z \leqslant O(k)$, so that $\theta = \max(z, k) = k$, and for small k it is less important than the terms of $O(k^{1/2})$ and $O(k^{P_{n+1}})$ which we keep. One can also see from Eq. (102) that there are no branch points for $z \leqslant O(k^2)$, since for z of this order the expression (103) is analytic in z. Consequently, in order to carry out the contour integral in Eq. (52), a consideration of $\delta U_{ij}^{\sigma-\sigma}$ requires that cuts be taken along two semiinfinite lines parallel to the negative part of the real axis, given by $-\infty \leqslant z \pm ick \leqslant 0$.

We now give an example of the type of branch points that appear in the contribution from two diffusive modes, $U_{ij}^{\lambda\mu}(\mathbf{k}, z)$. Consider the quantity $U_{\sigma\sigma}^{TT}(\mathbf{k}, z)$ which, according to (65), is given by

$$U_{\sigma\sigma}^{TT}(\mathbf{k}, z) = \frac{1}{2}\int' \frac{d\mathbf{q}}{(2\pi)^3}\,\frac{S_{\sigma\sigma}^{TT}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}})}{z + D_T(q^2 + l^2)} \tag{104}$$

where $S_{\sigma\sigma}^{TT}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}})$ is independent of $\hat{\mathbf{q}}$ and $\hat{\boldsymbol{l}}$, which follows from (C.13) and (C.18). Then

$$\begin{aligned} U_{\sigma\sigma}^{TT}(\mathbf{k}, z) &= \frac{1}{2} S_{\sigma\sigma}^{TT}\int \frac{d\mathbf{q}}{(2\pi)^3}\,[z + D_T(q^2 + l^2)]^{-1} \\ &= U_{\sigma\sigma}^{TT}(0, 0) - \frac{S_{\sigma\sigma}^{TT}(z + \frac{1}{2}D_T k^2)^{1/2}}{8\pi(2D_T)^{3/2}} + O(k) \end{aligned} \tag{105}$$

From Eq. (105) we see that for $z \sim O(k)$, there is a branch point singularity of the form $\sqrt{z}$ in $\delta U_{\sigma\sigma}^{TT}(\mathbf{k}, z)$, and one must cut the z plane along the line $-\infty \leqslant z \leqslant 0$. For $z \sim O(k^2)$, there is a branch point in the z plane at $z = -\frac{1}{2}D_T k^2$ and the cut extends from $-\infty \leqslant z \leqslant -\frac{1}{2}D_T k^2$. Finally, for $z < O(k^2)$ the function $\delta U_{\sigma\sigma}^{TT}(\mathbf{k}, z)$ is analytic in z. The analytic form of $\delta U_{ij}^{\lambda\mu}(\mathbf{k}, z)$ for diffusive mode contributions is, in general, more complicated than that given by Eq. (105). However, it can be shown by similar methods that $\delta U^{\lambda\mu}(\mathbf{k}, z)$ generally has a $\sqrt{z}$ singularity for $z = O(k)$, and for $z = O(k^2)$ there is a branch point at $z = -[D_\lambda D_\mu/(D_\lambda + D_\mu)]k^2$ and a cut extending from $-\infty \leqslant z \leqslant -[D_\lambda D_\mu/(D_\lambda + D_\mu)]k^2$, while for $z < O(k^2)$, $\delta U^{\lambda\mu}(\mathbf{k}, z)$ is analytic in z.

Although we will not give the details here, an examination of $\delta U^{\lambda\mu}(\mathbf{k}, z)$ and $\delta U^{\sigma\sigma}(\mathbf{k}, z)$ for branch points can be made along similar lines. The combinations of hydrodynamic modes represented by (λ, σ) and (σ, σ) give less singular contributions to $U(\mathbf{k}, z)$ than those coming from antiparallel sound modes, or from two diffusive modes. For example, for $z \sim O(k)$ and for $z \leqslant O(k^2)$, the function $\delta U^{\lambda\sigma}(\mathbf{k}, z)$ has a singularity of the form $z^2 \ln z$. In the following discussion we will see that singularities of these types are not as important for $\langle a_{\mathbf{k}}^i(t)\rangle$ as those contained in $\delta U^{\sigma-\sigma}(\mathbf{k}, z)$ or $\delta U^{\lambda\mu}(\mathbf{k}, z)$.

Next we address ourselves to questions (b) and (c). In order to discuss $\langle a_{\mathbf{k}}^i(t)\rangle$ for long times, we consider Eq. (52). To the order in k in which we have been consistent, the hydrodynamic matrix $\{z\mathbf{1} + ik\boldsymbol{\Omega}(\mathbf{k}) + k^2\mathbf{U}(\mathbf{k}, z)\}$ is diagonal, so that we can write

$$\langle a_{\mathbf{k}}^i(t)\rangle = \int_\Gamma \frac{dz\, e^{zt}}{2\pi i} \frac{\langle a_{\mathbf{k}}^i(0)\rangle}{z + ik\Omega_{ii}(\mathbf{k}) + k^2 U_{ii}(\mathbf{k}, z) + O(k^3)} \tag{106}$$

where the contour Γ is parallel to the imaginary axis and to the right of all poles and singularities of the integrand in (106). Consequently we may consider sound and diffusive modes separately. In general we may write a hydrodynamic mode $\langle a_{\mathbf{k}}^i(t)\rangle$ as a sum of two terms

$$\langle a_{\mathbf{k}}^i(t)\rangle = \langle a_{\mathbf{k}}^i(t)\rangle_p + \langle a_{\mathbf{k}}^i(t)\rangle_c \tag{107}$$

where $\langle a_{\mathbf{k}}^i(t)\rangle_p$ represents the contributions from the hydrodynamic poles, while $\langle a_{\mathbf{k}}^i(t)\rangle_c$ represents contributions from the cuts in the z plane. The terms $\langle a_{\mathbf{k}}^i(t)\rangle_p$ and $\langle a_{\mathbf{k}}^i(t)\rangle_c$ are given by expressions similar to (106) in which the contour Γ is replaced by contours Γ_p and Γ_c, respectively; the Γ_p are counterclockwise contours around the poles of the integrand in (106), and the Γ_c are counterclockwise contours around the cuts in the z plane.

We consider first a diffusive mode, $\langle a_{\mathbf{k}}^\eta(t)\rangle$ say, starting with the contributions from the poles. The poles are determined by Eq. (58), and are explicitly given by Eq. (89a). Now $\langle a_{\mathbf{k}}^\eta(t)\rangle_p$ is given by

$$\langle a_{\mathbf{k}}^\eta(t)\rangle_p = \exp\left[-t\left\{D_\eta k^2 - \sum_{n=1} \Delta_\eta(n) k^{2+P_n} + O(k^3)\right\}\right]\langle a_{\mathbf{k}}^\eta(0)\rangle \tag{108}$$

In order to be consistent with the fact that we have neglected terms of $O(k^3)$ in the hydrodynamic poles, we expand (108) and keep only the first two terms. That is,

$$\langle a_{\mathbf{k}}^\eta(t)\rangle_p = [\exp(-k^2 D_\eta t)]\left\{1 + k^2 t \sum_{n=1} \Delta_\eta(n) k^{P_n} + O[k(k^2 t)]\right\} \tag{109}$$

Consequently, on the scale of the damping time $\tau_d(k) = (D_\eta k^2)^{-1}$, the hydrodynamic poles give contributions of order $[t/\tau_d(k)]k^{P_n}$. We therefore infer that our results are only meaningful for the case where $t/\tau_d(k)$ is finite and where

k is small. If now we look at the contributions to $\langle a_{\mathbf{k}}{}^{\eta}(t)\rangle$ from the cuts, we write

$$\frac{\langle a_{\mathbf{k}}(t)\rangle_c}{\langle a_{\mathbf{k}}{}^{\eta}(0)\rangle} = \oint_{\Gamma_c} \frac{dz\, e^{zt}}{2\pi i} \left\{ z + k^2 D_\eta + \frac{1}{2} k^2 \sum_{ab} \delta U^{ab}_{\eta\eta}(\mathbf{k}, z) \right\}^{-1} \tag{110a}$$

$$= \oint_{\Gamma_c} \frac{dz\, e^{zt}}{2\pi i} \{z + D_\eta k^2\}^{-1}$$

$$- \frac{1}{2} k^2 \oint_{\Gamma_c} \frac{dz\, e^{zt}}{2\pi i} \frac{\sum_{ab} \delta U^{ab}_{\eta\eta}(\mathbf{k}, z)}{(z + D_\eta k^2)^2} + \cdots \tag{110b}$$

Here we have used Eq. (64b) for $U_{\eta\eta}(\mathbf{k}, z)$, and to be consistent with the terms that have been neglected, we expand the denominator in (110a) to obtain (110b). The first term in (110b) gives a vanishing contribution, since the integrand does not have any branch points. From the discussion of Eqs. (103)–(105) it follows that, on the scale of $\tau_d(k)$, the only important contributions to $\langle a_{\mathbf{k}}{}^{\eta}(t)\rangle_c$ come from combinations of diffusive modes.

One finds that the contribution to $\langle a_{\mathbf{k}}{}^{\eta}(t)\rangle_c$ coming from the cut in $\delta U^{\eta\eta}(\mathbf{k}, z)$ is of the order of $kf[\tau_d(k)/t] \exp(-\frac{1}{2}D_\eta k^2 t)$, where $f(x)$ is a bounded function of x for all values of x, and it depends on the detailed nature of the branch point. Consequently, for finite $t/\tau_d(k)$ and k small, the branch points make a contribution which is of the same order as those we have neglected in (109). However, to discuss the diffusive modes on the time scale of a sound oscillation, one should take the contribution from the cuts into account, as follows from (103) and (105), since they are of the same importance as those of the poles.

A similar analysis can be made of $\langle a_{\mathbf{k}}{}^{i}(t)\rangle_p$ and $\langle a_{\mathbf{k}}{}^{i}(t)\rangle_c$ for the other modes. In each case one finds that for times of the order of $\tau_d(k)$ the dominant contribution comes from the poles, and that for times of order $\tau_s(k)$ the contribution from the cuts must be included. A more detailed discussion of these points as they apply to the special case of self-diffusion will be given elsewhere.[13]

5. LONG-TIME TAILS AND CONVERGENCE CRITERIA

In this section we will discuss the long-time behavior of the time correlation functions, which are a direct consequence of the dispersion relations obtained in Section 3. We will further discuss the convergence of several expansions obtained here and in Section 3.

The mode–mode formula, together with the Navier–Stokes values of the hydrodynamic frequencies, yields the well-known $t^{-3/2}$ tails of the correlation functions, as shown in Ref. 5. We will show here that the higher corrections of $O(k^{2+P_n})$ (with $n \geqslant 1$ and $P_n = 1 - 2^{-n}$) to the dispersion relations (89) give rise to higher corrections to the $t^{-3/2}$ tails which are of $O(t^{-2P_{n+2}})$ with $n \geqslant 1$.

The time correlation functions $\tilde{C}_{ij}(0, t)$ are the inverse Laplace transforms of $C_{ij}(0, z)$, defined in (46), and $C_{ij}(0, z) = U_{ij}(0, z)$ according to (47). It follows then from Eq. (64b) that for small values of z

$$C_{ij}(0, z) = U_{ij} + \tfrac{1}{2} \sum_{a,b} \delta U_{ij}^{ab}(0, z) \tag{111}$$

By applying the results (B.7)–(B.11) of Appendix B, together with the full dispersion relations (89), one finds

$$C_{ij}(0, z) = U_{ij} - \sum_{\langle ab \rangle} \frac{M_{ij}^{ab}(0)}{4\pi\beta\rho D_{ab}^2} \sum_{n=0} \delta_{ab}(n) P_{n+2} \left(\frac{z}{2D_{ab}} \right)^{P_n+1} \tag{112}$$

where the first sum runs over the terms $\langle ab \rangle = (ss), (\eta, \eta), (\eta, T)$, and (T, T), we have used (98) and (99), and we introduced

$$\begin{aligned}
M_{ij}^{ss}(0) &= \frac{1}{2} \beta\rho \sum_{\sigma} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \\
M_{ij}^{\eta\eta}(0) &= \frac{1}{2} \beta\rho \sum_{m,n=1,2} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{\eta_m\eta_n}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \\
M_{ij}^{\eta T}(0) &= \beta\rho \sum_{m=1,2} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{\eta_m T}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \\
M_{ij}^{TT}(0) &= \frac{1}{2} \beta\rho \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{TT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})
\end{aligned} \tag{113}$$

where $(i, j) = (\eta_m, \sigma, l, T)$. The quantities $M_{ij}^{ab}(n)$, defined in Section 3, coincide with these definitions for the case $n = 0$. The coefficients $M_{lT}^{ab}(0)$, and therefore also $C_{lT}(0, z)$, vanish indentically, since the strength factors $S_{lT}^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ are either identically zero or odd functions in the angular variables [see (C.10)]. Equation (44) shows that the remaining off-diagonal correlation functions $C_{\sigma-\sigma}(0, z)$ and $C_{\sigma T}(0, z)$ are linear combinations of $C_{TT}(0, z)$ and $C_{ll}(0, z)$. We can therefore restrict ourselves to diagonal correlation functions $C_{ii}(0, z)$. Consequently, we only need the quantities $M_{ii}^{ab}(0)$, which are listed in Table I of Appendix C.

We now use the behavior of $C_{ii}(0, z)$ for small z to infer the behavior of its inverse Laplace transform $\tilde{C}_{ii}(0, t)$. To do this, we note that if a function $\tilde{f}(t)$ behaves like $\tilde{f}(t) \approx t^{-\mu}$, with $\mu > 1$ for large t, then its Laplace transform $f(z)$ behaves for small z as

$$f(z) \approx f(0) + \frac{\pi z^{\mu-1}}{\Gamma(\mu) \sin \pi\mu} \tag{114}$$

where $\Gamma(\mu)$ is the gamma function. Therefore the terms $z^{P_{n+1}}$ in expression (112) for $C_{ii}(0, z)$ yield terms $\sim t^{-(1+P_{n+1})}$ in $\tilde{C}_{ii}(0, t)$, so that

$$\tilde{C}_{ii}(0, t) \approx \sum_{\langle ab \rangle} \frac{M_{ii}^{ab}(0)}{\beta\rho(8\pi D_{ab}t)^{3/2}} \times \left\{1 + \sum_{n=1} \frac{\Delta_{ab}(n)}{D_{ab}} \frac{\Gamma(1 + 2P_{n+2})}{\Gamma(3/2)} \left(\frac{1}{2D_{ab}t}\right)^{P_n/2}\right\} \tag{115}$$

where the relations $\Gamma(\frac{1}{2}) = \sqrt{\pi}$ and $\Delta_{ab}(0) = 2D_{ab}/3$ have been used. We notice that all terms in the series are positive. Here again $i = (\eta, \sigma, l, T)$. The first term inside the curly brackets, which could have been included in the sum over n starting at $n = 0$, yields the well-known $t^{-3/2}$ tails. We further observe that the relative corrections to the $t^{-3/2}$ contribution of the mode pair (ab) do not depend on the correlation function considered, but only on the mode pair involved.

Another correlation function of physical interest which is not a particular case of Eq. (115) is the time correlation function $\tilde{C}_{\zeta\zeta}(0, t)$ for the bulk viscosity $\zeta = \rho D_l(0, 0) - \frac{4}{3}\eta(0, 0)$. The bulk viscosity is given in terms of the correlation function $C_{\zeta\zeta}(0, z)$ as $\zeta = \lim_{z\to 0} \rho C_{\zeta\zeta}(0, z)$. This correlation function can, of course, be introduced as

$$C_{\zeta\zeta}(0, z) = C_{ll}(0, z) - \tfrac{4}{3}C_{\eta\eta}(0, z) \tag{116}$$

and evaluated accordingly. Due to symmetry properties of isotropic tensors, $C_{\zeta\zeta}(0, z)$ can also be expressed in the more familiar form

$$C_{\zeta\zeta}(0, z) = \tfrac{1}{9}(\beta/\rho) \sum_{\alpha,\beta} (\hat{\tau}_{0\alpha\alpha}, \mathscr{G}_z \hat{\tau}_{0\beta\beta}) \tag{117}$$

where $(\alpha, \beta) = (x, y, z)$ label Cartesian components, and $\hat{\tau}_{0\alpha\alpha}$ is defined in (43b). Of course, the mode–mode integrals (64)–(67) apply also to this correlation function, and according to (67) and (32c) we have

$$\begin{aligned} A_\zeta^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) &= (\beta/\rho)^{1/2} \tfrac{1}{3} \sum_{x=x,y,z} (\hat{\tau}_{0xx}, a_{\mathbf{q}}{}^a a^b_{-\mathbf{q}}) \\ &= \tfrac{1}{3} \sum_{x=x,y,z} A_l^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \end{aligned} \tag{118}$$

Explicit expressions for A_ζ^{ab} are given in (A.21)–(A.24), and the corresponding strength factors, defined in (66), are calculated in (C.7)–(C.9).

The previous discussion allows us to extend Eqs. (112) and (113) to include the correlation function $C_{\zeta\zeta}(0, z)$. By comparing (112) with (89)–(96), we notice that in (112) all pairs of modes are involved [some of the coefficients $M_{ij}^{ab}(0)$ may of course still vanish], whereas in (89)–(96) this is only the case for the sound mode frequency while $z_\mu(k)$ involves only two opposite sound modes. This and other small differences are due to the fact that the mode–mode integrals are evaluated in different regions of k and z in each case.

In the second part of this section we investigate briefly the convergence of the series considered in Section 3 and in this section, using the convergence criterion that $\lim_{n\to\infty}(t_{n+1}/t_n) < 1$, where t_n is the nth term in any of these series. Since the convergence condition takes two different forms, depending on the series considered, we divide the series into two groups: (i) series (89a) for $z_\mu(k)$, the real part of series (89b) for $z_\sigma(k)$, and series (115) for $\tilde{C}_{ij}(0, t)$, and (ii) the imaginary part of series (89b) for $z_\sigma(k)$, series (100) and (101) for $\omega_s(x)$ and $\omega_\mu(x)$, respectively, and series (112) for $C_{ij}(0, z)$.

The convergence criterion imposes in case (i) the condition

$$\lim_{n\to\infty} \frac{\delta_s(n+1)}{\delta_s(n)} < 2 \tag{119a}$$

and in case (ii)

$$\lim_{n\to\infty} \frac{\delta_s(n+1)}{\delta_s(n)} < 1 \tag{119b}$$

To obtain these equations, we have used Eqs. (97)–(99) and the fact that $L_\mu^{ss}(n)$, defined in (93), approaches a constant, since it depends only on n through $P_n = 1 - 2^{-n}$.

To see whether (119a) and (119b) are indeed satisfied, we have to solve the recursion relations (97) for large values of n. For sufficiently large n, say $n \geqslant M$, the coefficients $L_\mu^{ss}(n)$ and $L_s^{ab}(n)$ in (97) approach their asymptotic values $L_\mu^{ss}(\infty)$ and $L_s^{ab}(\infty)$ arbitrarily closely, since they depend on n only through P_n, which can be checked from (93), (96), (76), and (82)–(85). For $n \geqslant M$ it is sufficient to replace the coefficients $L(n)$ in (97) by constants, and if we now eliminate $\delta_\mu(n)$ from (97a)–(97b), the recursion relation reduces to a second-order difference equation

$$\delta_s(n+2) - A\,\delta_s(n+1) - B\,\delta_s(n) = 0 \tag{120}$$

where A and B are positive quantities, defined as

$$A = L_s^{ss}(\infty) \tag{121a}$$

$$B = \sum_{\langle ab\rangle}{}' \tfrac{1}{2} L_s^{ab}(\infty)[L_a^{ss}(\infty) + L_b^{ss}(\infty)] \tag{121b}$$

The prime on the summation sign indicates that the sum runs only over the terms $\langle ab\rangle = (\eta\eta)$, (ηT), and (TT).

The solution of (120) is given by

$$\delta_s(n) = \alpha x_+{}^n + \beta x_-{}^n \tag{122}$$

where

$$x_\pm = \tfrac{1}{2}[A \pm (A^2 + 4B)^{1/2}] \tag{123}$$

Here α and β are constants. Since the positive root x_+ has a larger absolute value than x_-, we see that

$$\lim_{n\to\infty} \frac{\delta_s(n+1)}{\delta_s(n)} = x_+ \tag{124}$$

and the convergence conditions (119a) and (119b) require that x_+ be smaller than 1 or 2, respectively. By using (123) we have for the case (i) the convergence condition

$$A + B < 1 \tag{125}$$

and for case (ii)

$$2A + B < 4 \tag{126}$$

The coefficients A and B are expressed in terms of $L_{ab}(\infty)$, which are themselves proportional to $M^{ab}(\infty)$. These quantities are calculated explicitly in Appendix C, and are found as functions of transport coefficients and thermodynamic quantities, which can be measured experimentally. We have not made an attempt to analyze the available experimental data on fluids to check whether the convergence conditions are satisfied. However, for a dilute gas it is very simple to verify that (125) and (126) are satisfied. Inspection of the expressions (93) and (96) shows that all $L^{ab}(\infty)$ are proportional to the square of the reduced density $n^* = na^3$, where a is a measure of the range of the intermolecular forces. Hence (121) yields $A \sim (n^*)^2$ and $B \sim (n^*)^4$. In fact, using ideal gas values for all thermodynamic quantities, and estimating the viscosity η and the thermal conductivity λ by the first Enskog approximations for the transport coefficients of hard spheres with diameter a, i.e.,

$$\eta = \frac{5}{16a^2}\left(\frac{m}{\pi\beta}\right)^{1/2}; \qquad \lambda = \frac{15}{4}\frac{k_B}{m}\eta \tag{127}$$

we find that (125)–(126) reduce to $0.36(n^*)^2 + O(n^{*3}) < 1$ or 2, and this condition is satisfied for sufficiently small reduced densities. For the special case of hard spheres it seems possible to discuss the convergence of these series by calculating the quantities A and B in (121) by using the virial expressions for the equation of state and for the other thermodynamic quantities and by using Enskog's theory to estimate the transport coefficients for a hard sphere gas.

6. BURNETT AND SUPER-BURNETT EQUATIONS

In Section 3, and in Ref. 1, we showed that the hydrodynamic frequencies do not have analytic expansions in powers of k, but that terms proportional to $k^{5/2}$, $k^{11/4}$, etc. appear. This result has been established by assuming either the validity of the mode–mode formula, or, for a moderately dense gas, that the ring and repeated ring events give the dominant contributions. An important implication of this result is the observation that the linearized Burnett and super-Burnett corrections to the Navier–Stokes hydrodynamic equations do not properly describe the hydrodynamic behavior of the system, since the Burnett and super-Burnett corrections, if they exist, are at least of $O(k^3)$ and $O(k^4)$, respectively. One might suppose, therefore, that if one assumed *ab initio* that the hydrodynamic equation has a power series expression in k, some property of the Burnett and super-Burnett terms that result would indicate the presence of the $k^{5/2}$ term. In this section we show that this is indeed the case, and that the property referred to above is that the resulting Burnett, super-Burnett,..., transport coefficients are divergent.

We begin by considering the generalized hydrodynamic equations described in Section 2, in particular (30). We expect that the hydrodynamic description of the system will obtain for times long compared to some microscopic relaxation time, and we consider the long-time behavior of the generalized hydrodynamic equations. In such a case we neglect the term in (30) that involves the perpendicular part of the initial distribution function, $P_\perp \Phi(0)$, since this term is expected to decay rapidly compared to the terms which we retain, and we have

$$\sum_j \{z\,\delta_{ij} + ik\Omega_{ij}(k) + k^2 U_{ij}(\mathbf{k}, z)\}\langle a_{kz}^j\rangle = \langle a_\mathbf{k}^i(0)\rangle \tag{128}$$

It is convenient to transform (128) to the customary hydrodynamic variables: the density $\langle n_\mathbf{k}(t)\rangle$, the temperature $\langle T_\mathbf{k}(t)\rangle$, and the local velocity $\langle u_{\mathbf{k}\alpha}(t)\rangle$ ($\alpha = x, y, z$), where we have introduced the new microscopic functions

$$\begin{aligned} T_\mathbf{k}(\Gamma) &= \left(\frac{\partial T}{\partial e}\right)_n e_\mathbf{k}(\Gamma) + \left(\frac{\partial T}{\partial n}\right)_e n_\mathbf{k}(\Gamma) \\ u_{\mathbf{k}\alpha}(\Gamma) &= \frac{1}{\rho}\, g_{\mathbf{k}\alpha}(\Gamma), \qquad \alpha = x, y, z \end{aligned} \tag{129}$$

Since $(n_\mathbf{k}, T_\mathbf{k}) = 0$, as can be readily verified with the help of Appendix A, it follows that the functions $n_\mathbf{k}$, $T_\mathbf{k}$, and $u_\mathbf{k}$ ($\alpha = x, y, z$) form an orthogonal set of functions with respect to the inner product (13), and they can readily be normalized. Therefore the transformation of (128) to density, temperature, and local velocity is an orthogonal transformation of the old basis vectors in (17) to the new ones $\bar{a}_k^i$ ($i = 1, 2, 3, 4, 5$) with

$$\begin{gathered} \bar{a}_\mathbf{k}^1 = f_1^{-1} n_\mathbf{k}; \qquad \bar{a}_\mathbf{k}^2 = f_2^{-1} T_\mathbf{k} \\ \bar{a}_k^3 = f_3^{-1} u_{\mathbf{k}x}; \qquad \bar{a}_\mathbf{k}^4 = f_4^{-1} u_{\mathbf{k}y}; \qquad \bar{a}_k^5 = f_5^{-1} u_{\mathbf{k}z} \end{gathered} \tag{130}$$

The normalization constants are chosen so that

$$(\bar{a}_{\mathbf{k}}{}^i, \bar{a}_{\mathbf{k}}{}^j) = \delta_{ij} \tag{131}$$

which yields

$$\begin{aligned} f_1{}^2 &= (n_{\mathbf{k}}, n_{\mathbf{k}}) = n^2\kappa_T/\beta \\ f_2{}^2 &= (T_{\mathbf{k}}, T_{\mathbf{k}}) = T/\beta n C_V \\ f_3{}^2 = f_4{}^2 = f_5{}^2 &= (u_{\mathbf{k}x}, u_{\mathbf{k}x}) = 1/\beta\rho \end{aligned} \tag{132}$$

Here C_V is the specific heat per particle at constant volume, and $n\kappa_T = (\partial n/\partial P)_T$. The vector $\mathbf{k}$ is taken parallel to the x axis. Equation (132) can be verified with the help of Appendix A.

In the new representation (130), the hydrodynamic equations (128) read

$$\sum_j \{z\, \delta_{ij} + ik\bar{\Omega}_{ij}(\mathbf{k}) + k^2\bar{U}_{ij}(\mathbf{k}, z)\}\langle \bar{a}_{\mathbf{k}z}^j\rangle = \langle \bar{a}_{\mathbf{k}}{}^i(0)\rangle \tag{133}$$

Instead of carrying out the orthogonal transformation to obtain $\bar{\Omega}_{ij}$ and $\bar{U}_{ij}$ from Ω_{ij} and U_{ij}, respectively, it is simpler to evaluate $\bar{\Omega}_{ij}$ and $\bar{U}_{ij}$ directly from Eqs. (24) and (25), in which the $a_{\mathbf{k}}{}^i$ are replaced by $\bar{a}_{\mathbf{k}}{}^i$. The result is

$$\begin{aligned} &\bar{\Omega}_{13} = \bar{\Omega}_{31} = c\gamma^{-1/2}, \qquad \bar{\Omega}_{23} = \bar{\Omega}_{32} = c[(\gamma - 1)/\gamma]^{1/2} \\ &\bar{\Omega}_{ij} = 0 \text{ in all other cases} \end{aligned} \tag{134}$$

and

$$\begin{aligned} \bar{U}_{22}(\mathbf{k}, z) &= (nC_V)^{-1}\lambda(\mathbf{k}, z) \\ \bar{U}_{33}(\mathbf{k}, z) &= D_l(\mathbf{k}, z) \\ \bar{U}_{44}(\mathbf{k}, z) &= U_{55}(\mathbf{k}, z) = \rho^{-1}\eta(\mathbf{k}, z) \\ \bar{U}_{23}(\mathbf{k}, z) &= U_{32}(\mathbf{k}, z) = (\rho n T C_V)^{-1/2}\theta(\mathbf{k}, z) \\ \bar{U}_{ij}(\mathbf{k}, z) &= 0 \text{ in all other cases} \end{aligned} \tag{135}$$

The symbols are defined in (43)–(45). Before proceeding, we want to remark that the hydrodynamic equation (133) could have been used in Section 2 just as well as those given by (128). The roots of the secular equations of (128) and (133) are the same, and are determined by (56)–(58). The advantage of the representation (17) is that it diagonalizes the matrix $\mathbf{\Omega}(k)$ to lowest order in k.

Equation (133) represents the Fourier–Laplace transforms of the usual hydrodynamic equations, which we will write out explicitly in terms of the variables $\langle n_{\mathbf{k}}(t)\rangle$, $\langle T_{\mathbf{k}}(t)\rangle$, and $\langle \mathbf{u}_{\mathbf{k}}(t)\rangle$. This can be done most easily by replacing $\bar{a}_{\mathbf{k}}^j$ by $f_j\bar{a}_{\mathbf{k}}^j$, $\bar{\Omega}_{ij}$ by $f_i f_j^{-1}\bar{\Omega}_{ij}$, and $\bar{U}_{ij}$ by $f_i f_j^{-1}\bar{U}_{ij}$, and inverting the Laplace transforms,

$$\begin{aligned} \partial_t\langle n_k(t)\rangle &= -ikn\langle u_{\mathbf{k}x}(t)\rangle \\ \partial_t\langle T_{\mathbf{k}}(t)\rangle &= -ik\alpha^{-1}(\gamma-1)\langle u_{\mathbf{k}x}(t)\rangle - ik(nC_V)^{-1}\langle q_{\mathbf{k}x}(t)\rangle \\ \partial_t\langle u_{\mathbf{k}\alpha}(t)\rangle &= -ik\rho^{-1}\langle P_{\mathbf{k}x\alpha}(t)\rangle, \qquad \alpha = x, y, z \end{aligned} \tag{136}$$

We have made the following identifications for the longitudinal component of the heat current $\langle q_{\mathbf{k}x}(t)\rangle$ and the longitudinal components of the pressure tensor $\langle P_{\mathbf{k}x\alpha}(t)\rangle$:

$$\begin{aligned} \langle q_{\mathbf{k}x}(t)\rangle = &-ik\int_0^t d\tau\, \tilde{\lambda}(\mathbf{k},\tau)\langle T_{\mathbf{k}}(t-\tau)\rangle \\ &-ik\int_0^t d\tau\, \tilde{\theta}(\mathbf{k},\tau)\langle u_{\mathbf{k}x}(t-\tau)\rangle \end{aligned} \tag{137a}$$

$$\begin{aligned} \langle P_{\mathbf{k}xx}(t)\rangle = \langle P_{\mathbf{k}}(t)\rangle &- ik\int_0^t d\tau\, \rho\tilde{D}_l(\mathbf{k},\tau)\langle u_{\mathbf{k}x}(t-\tau)\rangle \\ &- \frac{ik}{T}\int_0^t d\tau\, \tilde{\theta}(\mathbf{k},\tau)\langle T_{\mathbf{k}}(t-\tau)\rangle \end{aligned} \tag{137b}$$

$$\langle P_{\mathbf{k}xy}\rangle = -ik\int_0^t d\tau\, \tilde{\eta}(\mathbf{k},\tau)\langle u_{\mathbf{k}y}(t-\tau)\rangle \tag{137c}$$

and the local pressure $\langle P_{\mathbf{k}}(t)\rangle$ is found here as

$$\langle P_{\mathbf{k}}(t)\rangle = \left(\frac{\partial P}{\partial n}\right)_T\langle n_{\mathbf{k}}(t)\rangle + \left(\frac{\partial P}{\partial T}\right)_n\langle T_{\mathbf{k}}(t)\rangle \tag{138}$$

where we have used the thermodynamic relations

$$c\gamma^{-1/2}f_3f_1^{-1} = \frac{1}{\rho}\left(\frac{\partial P}{\partial n}\right)_T; \qquad c\left(\frac{\gamma-1}{\gamma}\right)^{1/2}f_3f_2^{-1} = \frac{1}{\rho}\left(\frac{\partial P}{\partial T}\right)_n \tag{139}$$

In general, $\tilde{F}(t)$ indicates the inverse Laplace transform of $F(z)$. In the limit of large t and small k, (137a)–(137c) reduce to the usual linear relations between the fluxes and the gradients, since the function $\tilde{\theta}(k, t)$ vanishes in this limit, as we have seen in Section 2, and the time variation over the time τ of the hydrodynamic variables $\langle T_{\mathbf{k}}(t-\tau)\rangle$ and $\langle \mathbf{u}_{\mathbf{k}}(t-\tau)\rangle$ may be neglected, since it is of higher order in k, as follows from (136). Finally, the time integrals over $(0, \infty)$ of $\tilde{\lambda}(0, \tau)$, $\tilde{D}_l(0, \tau)$, and $\tilde{\eta}(0, \tau)$ are the time correlation function expressions for the Navier–Stokes transport coefficients λ, D_l, and η, respectively. The quantity $\tilde{\theta}(k, \tau)$ is a transport coefficient which couples the heat flow and the longitudinal momentum current, and does not appear in the linearized Navier–Stokes equations.

We may obtain the hydrodynamic equations to order k^3 by making a

k-expansion of the quantities $\tilde{\lambda}(\mathbf{k}, t)$, $\tilde{\theta}(\mathbf{k}, t)$, $\tilde{D}_l(\mathbf{k}, t)$, and $\tilde{\eta}(\mathbf{k}, t)$, and a τ-expansion of $\langle T_\mathbf{k}(t - \tau)\rangle$ and $\langle \mathbf{u}_\mathbf{k}(t - \tau)\rangle$. The latter yields

$$\begin{aligned}\langle T_\mathbf{k}(t - \tau)\rangle &= \langle T_\mathbf{k}(t)\rangle - \tau\, \partial_t \langle T_\mathbf{k}(t)\rangle + \cdots \\ &= \langle T_\mathbf{k}(t)\rangle + ik\tau \frac{\gamma - 1}{\alpha} \langle u_{\mathbf{k}x}(t)\rangle + O(k^2) \end{aligned} \tag{140a}$$

$$\langle u_{\mathbf{k}x}(t - \tau)\rangle = \langle u_{\mathbf{k}x}(t)\rangle + ik\tau \frac{1}{\rho} \langle P_\mathbf{k}(t)\rangle + O(k^2) \tag{140b}$$

$$\langle u_{\mathbf{k}y}(t - \tau)\rangle = \langle u_{\mathbf{k}y}(t)\rangle - (ik)^2 \tau \frac{\eta}{\rho} \langle u_{\mathbf{k}y}(t)\rangle + O(k^4) \tag{140c}$$

We have used the hydrodynamic equations (136) and (137) to lowest non-vanishing order to eliminate the time derivatives in favor of the k-expansion. Next we consider the functions $\tilde{\lambda}$, $\tilde{D}_l$, $\tilde{\eta}$, and $\tilde{\theta}$, defined in (45). By examining these expressions under translations, rotations, and reflections in configuration and velocity space, one finds that the k-expansions of $\tilde{\lambda}$, $\tilde{D}_l$, and $\tilde{\eta}$ contain only even powers of k, whereas $\tilde{\theta}$ contains only odd powers of k, i.e.,

$$\begin{aligned}\tilde{\lambda}(\mathbf{k}, t) &= \tilde{\lambda}_0(t) + (ik)^2 \tilde{\lambda}_2(t) + O(k^4) \\ \tilde{D}_l(\mathbf{k}, t) &= \tilde{D}_{l0}(t) + (ik)^2 \tilde{D}_{l2}(t) + O(k^4) \\ \tilde{\eta}(\mathbf{k}, t) &= \tilde{\eta}_0(t) + (ik)^2 \tilde{\eta}_2(t) + O(k^4) \\ \tilde{\theta}(\mathbf{k}, t) &= ik\tilde{\theta}_1(t) + (ik)^3 \tilde{\theta}_3(t) + O(k^5)\end{aligned} \tag{141}$$

As a result of these expansions, we may make a k-expansion of the heat current and the longitudinal components of the pressure tensor as

$$\begin{aligned}\langle q_{\mathbf{k}x}(t)\rangle = &-ik\lambda(t)\langle T_\mathbf{k}(t)\rangle - (ik)^2 \frac{\gamma - 1}{\alpha} \vartheta_2(t) \langle u_{\mathbf{k}x}(t)\rangle \\ &+ (ik)^2 \tfrac{2}{3} \vartheta_4(t) \langle u_{\mathbf{k}x}(t)\rangle + O(k^3) \end{aligned} \tag{142a}$$

$$\begin{aligned}\langle P_{\mathbf{k}xx}(t)\rangle = &\langle P_\mathbf{k}(t)\rangle - ik\rho D_l(t) \langle u_{\mathbf{k}x}(t)\rangle \\ &- \frac{2}{3}\frac{1}{\rho}\, \omega_2(t)(ik)^2 \langle P_\mathbf{k}(t)\rangle + \frac{2}{3}\, \omega_3(t)(ik)^2 \langle T_\mathbf{k}(t)\rangle + O(k^3)\end{aligned} \tag{142b}$$

$$\langle P_{\mathbf{k}xy}(t)\rangle = -ik\eta(t)\langle u_{\mathbf{k}y}(t)\rangle + (ik)^3 \omega(t) \langle u_{\mathbf{k}y}(t)\rangle + O(k^5) \tag{142c}$$

The (time-dependent) Navier–Stokes transport coefficients are given by

$$\begin{aligned}\lambda(t) &= \int_0^t d\tau\, \tilde{\lambda}_0(\tau), \\ D_l(t) &= \int_0^t d\tau\, \tilde{D}_{l,0}(\tau) \\ \eta(t) &= \int_0^t d\tau\, \tilde{\eta}_0(\tau)\end{aligned} \tag{143}$$

The (time-dependent) Burnett coefficients are given by

$$\begin{aligned}
\vartheta_2(t) &= \int_0^t d\tau\, \tau\tilde{\lambda}_0(\tau), \qquad \vartheta_4(t) = -\tfrac{3}{2}\int_0^t d\tau\, \theta_1(\tau) \\
\omega_2(t) &= \tfrac{3}{2}\int_0^t d\tau\, \tau\rho\tilde{D}_{l,0}(\tau) \\
\omega_3(t) &= -\tfrac{3}{2}\,(1/T)\int_0^t d\tau\, \tilde{\theta}_1(\tau) = (1/T)\vartheta_4(t)
\end{aligned} \tag{144}$$

and the (time-dependent) super-Burnett coefficient is given by

$$\omega(t) = -\int_0^t d\tau\, \tilde{\eta}_2(\tau) + \frac{\eta}{\rho}\int_0^t d\tau\, \tau\tilde{\eta}_0(\tau) \tag{145}$$

Here we have made an attempt to keep our notation consistent with that used by Wang Chang and Uhlenbeck,[(14)] and partially by Chapman and Cowling,[(15),5] for the case of the dilute gas. If, for sufficiently long times, these transport coefficients approach constant values, then substitution of (142) into equation (137) will lead to the linear Burnett and super-Burnett (for the case of the transverse velocity field) equations with constant transport coefficients. Such equations would then lead to hydrodynamic frequencies $z(k)$, which have well-defined expansions in powers of k, where the coefficients are determined to order k^2 by $\lambda(\infty)$, $D_l(\infty)$, and $\eta(\infty)$; to order k^3 by $\vartheta_2(\infty)$, $\vartheta_4(\infty)$, $\omega_2(\infty)$, and $\omega_3(\infty)$; and to order k^4 for the transverse velocity field by $\omega(\infty)$.

This picture cannot be maintained, however, since both the mode–mode coupling theory and the kinetic of gases have suggested that $\tilde{\lambda}_0(\tau)$, $\tilde{D}_{l,0}(\tau)$, and $\tilde{\eta}_0(\tau)$ depend on τ for large τ as $\tau^{-3/2}$. Such a conclusion is based on the two-mode approximation used in this paper, or on the ring events used in Ref. 1. As a result of this $\tau^{-3/2}$ behavior one can see that, although $\lambda(\infty)$, $D_l(\infty)$, and $\eta(\infty)$ exist, the Burnett and super-Burnett transport coefficients $\vartheta_2(t)$, $\omega_2(t)$, and $\omega(t)$ do not exist as $t \to \infty$. Let us investigate the precise behavior of all Burnett and super-Burnett coefficients in a systematic way. From (44) we deduce that $\lambda(\mathbf{k}, z)$, $D_l(\mathbf{k}, z)$, $\eta(\mathbf{k}, z)$, and $\theta(\mathbf{k}, z)$ are related to $U_{ij}(\mathbf{k}, z)$ as follows:

$$\begin{aligned}
\lambda(\mathbf{k}, z) &= nC_P U_{TT}(\mathbf{k}, z); \qquad D_l(\mathbf{k}, z) = U_{ll}(\mathbf{k}, z) \\
\eta(\mathbf{k}, z) &= \rho U_{\eta_1\eta_1}(\mathbf{k}, z); \qquad \theta(\mathbf{k}, z) = \frac{c\rho\alpha T}{(\gamma - 1)^{1/2}}\, U_{Tl}(\mathbf{k}, z)
\end{aligned} \tag{146}$$

[5] To compare the notation used in this paper with that of Chapman and Cowling, we note that using their Eqs. (15.3.4) and (15.3.6) and our Eqs. (144) and (145), the following relations hold:

$$\vartheta_2 = \theta_2\mu^2/\rho T; \qquad \vartheta_4 = \theta_4\mu^2/\rho;$$
$$\omega_2 = \bar{\omega}_2\mu^2/P; \qquad \omega_3 = \bar{\omega}_3\mu^2/\rho T$$

We will therefore consider the k-expansion of $\tilde{U}_{ij}(\mathbf{k}, t)$ in general. In fact, we are only interested in the dominant contributions to these terms for large t and small k, or equivalently in the leading singularities in $U_{ij}(\mathbf{k}, z)$ for small k and z. According to our basic assumption, the leading singularities in $U_{ij}(\mathbf{k}, z)$ are given by the mode–mode integrals, which are analyzed in Appendix B. It is found there that for small k and z

$$U_{ij}(\mathbf{k}, z) \simeq U_{ij}(0, 0) - \frac{1}{2}\sum_{\lambda\mu}\int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{\lambda\mu}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \frac{z^{1/2}}{4\pi(2D_{\lambda\mu})^{3/2}}$$
$$- \frac{1}{2}\sum_{\sigma}\int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \frac{(z + i\sigma ck\hat{q}_x)^{1/2}}{4\pi(2D_s)^{3/2}} \tag{147}$$

where the sum over λ and μ runs over the diffusive modes (η_1, η_2, T), and σ runs over the sound modes ($\sigma = \pm$). A straightforward k-expansion of (147) at fixed z and subsequent inversion of the Laplace transform by means of (114) yields

$$\tilde{U}_{ij}(k, t) = \tilde{U}_{0,ij}(t) + ik\tilde{U}_{1,ij}(t) + (ik)^2\tilde{U}_{2,ij}(t) + \cdots \tag{148}$$

where the coefficients for large t are given by

$$\tilde{U}_{0,ij}(t) \simeq t^{-3/2} \sum_{\langle ab\rangle} \frac{M_{ij}^{ab}(0)}{\beta\rho(8\pi D_{ab})^{3/2}} \tag{149a}$$

$$\tilde{U}_{1,ij}(t) \simeq \frac{-ct^{-1/2}}{(8\pi D_s)^{3/2}} \frac{1}{2}\sum_{\sigma}\int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})\sigma\hat{q}_x \tag{149b}$$

$$\tilde{U}_{2,ij}(t) \simeq \frac{c^2t^{1/2}}{2(8\pi D_s)^{3/2}} \frac{1}{2}\sum_{\sigma}\int \frac{d\hat{\mathbf{q}}}{4\pi} S_{ij}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})\hat{q}_x^{\,2} \tag{149c}$$

In (149a) we have introduced the matrix elements $M_{ij}^{ab}(0)$, which are defined in (113) and have the values given in Appendix C. The summation index $\langle ab\rangle$ takes the values $(\eta\eta)$, (ηT), (TT), and (ss), and $D_{ab} = \frac{1}{2}(D_a + D_b)$. Equation (149a) describes the long-time behavior of the correlation functions, and is in fact the first term of (115). The matrix elements of $\tilde{U}_0(t)$ and $\tilde{U}_2(t)$, or $\tilde{U}_1(t)$, are only nonvanishing when $S_{ij}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ contains terms which are respectively, even or odd in $\hat{q}_x$.

If we apply (149a)–(149c) to (146), we see from the expressions (C.4)–(C.10) in Appendix C that $S_{TT}^{\sigma-\sigma}$, $S_{ll}^{\sigma-\sigma}$, and $S_{\eta_1\eta_1}^{\sigma-\sigma}$ are even functions of $\hat{q}_x$, and $S_{lT}^{\sigma-\sigma}$ is an odd function of $\hat{q}_x$. Therefore the k-expansions of the functions $\tilde{\lambda}(\mathbf{k}, t)$, $\tilde{D}_l(\mathbf{k}, t)$, and $\tilde{\eta}(\mathbf{k}, t)$ contain only even powers of k, where $\tilde{\lambda}_0(t)$, $\tilde{D}_{l,0}(t)$, and $\tilde{\eta}_0(t)$, defined in (141), can be taken directly from (149a) and

(146). We further need to know $\tilde{\eta}_2(t)$, defined in (141). According to (146) and (149c), it is given by

$$\tilde{\eta}_2(t) \simeq \rho \frac{c^2 t^{1/2}}{2(8\pi D_s)^{3/2}} \frac{1}{2} \sum_\sigma \int \frac{d\hat{\mathbf{q}}}{4\pi} S^{\sigma-\sigma}_{\eta_1\eta_1}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})\hat{q}_x{}^2$$

$$= \frac{c^2 t^{1/2}}{70\beta(8\pi D_s)^{3/2}} \tag{150}$$

where we have used (C.4). The k-expansion of $\tilde{\theta}(\mathbf{k}, t)$ contains only odd powers of k, and, according to (141), (146), and (149b), is given by

$$\tilde{\theta}_1(t) \simeq -\frac{\rho c\alpha T}{(\gamma-1)^{1/2}} \frac{ct^{-1/2}}{(8\pi D_s)^{3/2}} \frac{1}{2} \sum_\sigma \int \frac{d\hat{\mathbf{q}}}{4\pi} S^{\sigma-\sigma}_{lT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})\sigma\hat{q}_x$$

$$= -\frac{c^2 t^{-1/2}}{3\beta(8\pi D_s)^{3/2}} \left\{\frac{3}{5} - \frac{\gamma-1}{\alpha T} + \frac{n}{c}\left(\frac{\partial c}{\partial n}\right)_s\right\} \tag{151}$$

where we have used (C.10) and (C.18).

From Eqs. (148)–(151) we can determine the precise behavior of the Burnett and super-Burnett coefficients defined in (144) and (145). The results for the Burnett coefficients are

$$\vartheta_2(t) \simeq \frac{2C_P}{\beta m} \sum_{\langle ab\rangle} \frac{M^{ab}_{TT}(0)}{(8\pi D_{ab})^{3/2}} t^{1/2} \tag{152a}$$

$$\vartheta_4(t) = T\omega_3(t) \approx \frac{c^2}{\beta(8\pi D_s)^{3/2}} \left\{\frac{3}{5} - \frac{\gamma-1}{\alpha T} + \frac{n}{c}\left(\frac{\partial c}{\partial n}\right)_s\right\} t^{1/2} \tag{152b}$$

$$\omega_2(t) \simeq \frac{3}{\beta} \sum_{\langle ab\rangle} \frac{M^{ab}_{ll}(0)}{(8\pi D_{ab})^{3/2}} t^{1/2} \tag{152c}$$

and for the super-Burnett coefficient

$$\omega(t) \simeq \frac{-c^2}{105\beta(8\pi D_s)^{3/2}} t^{3/2} \tag{153}$$

The values of the dimensionless coefficients $M^{ab}_{ij}(0)$ are given in Appendix C.

We have shown, therefore, that all Burnett coefficients $\vartheta_2(t)$, $\vartheta_4(t)$, $\omega_2(t)$, and $\omega_3(t)$ grow as $t^{1/2}$ as t gets large, and that the super-Burnett coefficient grows as $t^{3/2}$ as t gets large. Notice also that the second term in (145) does not contribute to the dominant time behavior. In addition, one can see by inspecting the expressions for the heat flux and the pressure tensor that the coefficients of the $t^{1/2}$ term in these quantities do not vanish by a fortuitous cancellation.

7. DISCUSSION

In this paper we have considered the analytic structure of the linearized hydrodynamic equations. Our analysis has been based either (for a general fluid without internal degrees of freedom) on the application of the mode–mode coupling theory to the elements of the transport matrix $U_{ij}(\mathbf{k}, z)$, or (as discussed in Ref. 1 for a gas of hard spheres at moderate densities) on the assumption that the behavior of $U_{ij}(\mathbf{k}, z)$ for small k and z is determined by the ring (and repeated ring) dynamical events.

The major results of the analysis carried out here are as follows.

1. The hydrodynamic equations beyond the Navier–Stokes order have a very intricate structure. There is no longer an orderly progression from the order-k^2 Navier–Stokes equations, to the order-k^3 Burnett equations, to the order-k^4 super-Burnett equations, and so on, as one considers disturbances of increasingly larger wave number k. Instead, there is an infinite number of powers, of the form k^{2+P_n}, $P_n = 1 - 2^{-n}$, $n = 1, 2, \ldots$, which appear between order k^2 and order k^3. This is because the dispersion relations have a k-expansion of this form.
2. In addition to poles, the hydrodynamic matrix also has branch points. The relative importance of the pole and branch point contributions depends on the particular time scale of interest. We found that for times of order $\tau_d(k) \approx (Dk^2)^{-1}$ the k^{2+P_n} corrections to the hydrodynamic poles are more important than the contributions from the cuts in the z plane, but that for times of order $\tau_s(k) \sim (ck)^{-1}$ the branch point contributions are at least as important as the k^{2+P_n} corrections to the poles.
3. Asymptotic correction terms to the long-time behavior of the correlation functions, which are of the form $t^{-2P_{n+2}}$ with $n = 1, 2, 3, \ldots$, are a direct consequence of the k^{2+P_n} terms in the dispersion relations.
4. A straightforward expansion of the hydrodynamic equations in powers of k leads to expressions for the Burnett and super-Burnett transport coefficients which diverge in the long-time limit. In this paper we have considered the coefficients connected with the hydrodynamic equations for the velocity and temperature.

It is interesting to compare these results for the generalized hydrodynamic equations in fluids with the generalization of Fick's diffusion equation to higher order in k. To find corrections to Fick's law, one can again analyze the transport matrix elements by means of the mode–mode formula, and can determine the hydrodynamic poles and the location and nature of the branch point singularities. The dispersion relation for the relaxation time of the diffusive mode does not contain terms of the form k^{2+P_n}, $n = 1, 2, \ldots$. This is because the antiparallel sound modes, which are solely responsible for the k^{2+P_n} terms in the frequencies of diffusive modes (heat and shear), do not contribute in the self-diffusion case. In fact, it appears that it is not possible to describe the decay of an excess density of tagged particles in terms of corrections to Fick's law in the form of a dispersion relation, since on the time scale $\tau_d(k) \sim (Dk^2)^{-1}$ the contributions from branch cuts are more important than the contributions from the hydrodynamic poles.[13]

The long-time behavior of the velocity autocorrelation function $\langle v_x(t)v_x\rangle_{\text{eq}}$ shows the same behavior as the correlation functions discussed in (115), and its dominant contribution for long times comes from a combination of a shear mode and the self-diffusion mode:

$$\langle v_x(t)v_x\rangle_{\text{eq}} \simeq \frac{2}{3\beta\rho[4\pi(D_\eta + D)t]^{3/2}} \times \left\{1 + \sum_n \frac{\Delta_\eta(n)}{D_\eta + D} \frac{\Gamma(1 + 2P_{n+2})}{\Gamma(3/2)} [(D_\eta + D)t]^{-P_n/2}\right\}$$

where D is the coefficient of self-diffusion. We have used the fact that the frequency of the self-diffusion mode does not contain terms of $O(k^{2+P_n})$.

The coefficient of the $t^{-3/2}$ tail has here been taken from Ref. 5. The difference between diffusive modes in fluids and the diffusive mode of tagged particles is also reflected in the behavior of the super-Burnett coefficient, as has been discussed by Keyes and Oppenheim,[16] Dufty and McLennan,[17] and de Schepper *et al.*[18] In the case of self-diffusion, the appropriate super-Burnett coefficient diverges as $t^{1/2}$ for large t, not as $t^{3/2}$ as is typical for the super-Burnett coefficient discussed in Section 6. This is because combinations of antiparallel sound modes, which lead to the $t^{3/2}$ divergence for other transport processes, do not contribute to the super-Burnett coefficient for self-diffusion.

Results similar to those given here for the coefficients of the k^{2+P_n} terms have been found by Pomeau.[6] However, in almost every case the coefficients $\Delta_\mu(n)$, $\Delta_s(n)$, and $\overline{\Delta}_s(n)$, given by Eqs. (89)–(99) differ from the corresponding results found by Pomeau. This difference can be traced to four main sources of error in Pomeau's calculations: (i) Trivial mistakes either in sign or in algebra. (ii) A systematic replacement of the quantity which corresponds to $S(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ in our Section 3 by an average value $\overline{S}$, where $\overline{S} = (4\pi)^{-1}\int d\hat{\mathbf{q}}\, S(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$. This replacement is not correct in the mode–mode formula for $U(\mathbf{k}, z)$ given by Eq. (65), since denominators appear which also have a dependence on the $\hat{\mathbf{q}}$. This wrong approximation also explains why Pomeau's coefficients for the long-time tails in $\tilde{C}_{ii}(0, t)$, defined by Eq. (115), and his coefficients for the k^{2+P_n} terms in the dispersion relations (89) are related in a simple way. However, it is clear that in $\tilde{C}_{ii}(0, t)$ a replacement of $S(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ by $\overline{S}$ is allowed, since nothing else in the appropriate integral depends on $\hat{\mathbf{q}}$, while it is not allowed in the calculations of $U(\mathbf{k}, z)$. (iii) The term $\theta(\mathbf{k}, z)$ is neglected in the expression for the sound damping constant

$$U_{\sigma\sigma}(\mathbf{k}, z) = \tfrac{1}{2}D_l(\mathbf{k}, z) + \tfrac{1}{2}(\gamma - 1)D_T(\mathbf{k}, z) + \sigma\theta(\mathbf{k}, z)(\alpha c/nCP)$$

Pomeau has argued that $\theta(\mathbf{k}, z)$ is proportional to k, while here we have demonstrated that the term is of $O(k^{1/2})$. (iv) The hydrodynamic frequencies are determined as the solution of the wrong equation. That is, Pomeau solves $z_\mu = k^2 D_\mu(\mathbf{k}, z_\mu)$ and $z_\sigma = i\sigma ck + k^2 u_{\sigma\sigma}(\mathbf{k}, z)$, instead of the correct equations as given by Eqs. (55)–(57).

It is of considerable interest to determine whether the complex analytic structure found for the hydrodynamic equations has any relevance for experiment. In Ref. 1 we showed that, for dilute gases, the $k^{5/2}$ terms present in the sound dispersion equation are undetectable at the present time. It might be more appropriate to look for applications of the results obtained here, which hold for a general fluid, to experiments on fluids at liquid densities. It remains to analyze the results given here in order to determine the magnitude of the effects in experimental situations.

APPENDIX A

In this appendix we will briefly review those fluctuation formulas that are needed in the body of the paper, and which have been evaluated by Ernst *et al.*[5] We refer to their paper for the details of the calculations. In this appendix, expressions for the mode–mode amplitudes $A_i^{rs}(\hat{\mathbf{q}}, \mathbf{l})$ are also given.

We consider fluctuation formulas in a grand canonical ensemble, characterized by the parameters $\beta = (k_B T)^{-1}$ and $\nu = \beta\mu$, where μ is the chemical potential. The fluctuation formulas considered are given in general in the form $(a_\mathbf{k}, b_\mathbf{k})$ and $(a_\mathbf{k}, b_\mathbf{q} c_\mathbf{l})$ where the inner product is defined in Eq. (13), and where $a_\mathbf{k}$, $b_\mathbf{k}$, and $c_\mathbf{k}$ are Fourier transforms of microscopic densities which satisfy $(1, a_\mathbf{k}) = (1, b_\mathbf{k}) = (1, c_\mathbf{k}) = 0$.

We consider first $(a_\mathbf{k}, b_\mathbf{k})$ and observe that for small values of k the inner products have the form

$$(a_\mathbf{k}, b_\mathbf{k}) = (a_0, b_0) + O(k^2) \tag{A.1}$$

when $a_\mathbf{k}$ and $b_\mathbf{k}$ are scalar functions. The term linear in k vanishes due to spatial isotropy of the equilibrium averages. Now let $a_\mathbf{k}$ be the Fourier transform of one of the microscopic functions $n_\mathbf{k}$, $\mathbf{g}_\mathbf{k}$, $e_\mathbf{k}$, or $\tau_{\mathbf{k}xx}$, defined in Eqs. (10) and (34). In the small-k limit they satisfy the relations

$$(a_0, e_0) = -(\partial a/\partial\beta)_\nu; \qquad (a_0, n_0) = +(\partial a/\partial\nu)_\beta \tag{A.2a}$$

$$(a_0, S_0) = -(nT)^{-1}(\partial a/\partial\beta)_P; \qquad (a_0, P_0) = n\beta^{-1}(\partial a/\partial n)_S \tag{A.2b}$$

where $S_{\mathbf{k}}$ and $P_{\mathbf{k}}$ are defined in (15) and where n, e, P, and S are, respectively, the values of the number density, energy density, pressure, and entropy per particle in thermal equilibrium. Here a_0 should, according to Eq. (9), be taken as

$$a_0 = A - \langle A \rangle_{\mathrm{eq}} \tag{A.3}$$

where $A = \int d\mathbf{r}\, a(\mathbf{r})$ and $a = \langle a(\mathbf{r}) \rangle_{\mathrm{eq}} = V^{-1} \langle A \rangle_{\mathrm{eq}}$. For $a = n, e$, and $\mathbf{g}$ these expressions are given in Eq. (10a). For the longitudinal component of the longitudinal momentum density one has from Eqs. (34) and (9)

$$\tau_{0xx} = J_{xx} - \langle J_{xx} \rangle_{\mathrm{eq}} = J_{xx} - PV \tag{A.4}$$

where, according to (34),

$$J_{xx} = \sum_{i=1}^{N} m v_{ix}^2 - \frac{1}{2} \sum_{i \neq j=1}^{N} \sum^{N} r_{ij,x} \frac{\partial \phi(r_{ij})}{\partial r_{ij,x}} \tag{A.5}$$

The second equality in (A.4) is the virial theorem.

If $a_{\mathbf{k}}$ in (A.2) is a linear combination of $n_{\mathbf{k}}$, $e_{\mathbf{k}}$, $\mathbf{g}_{\mathbf{k}}$, and $\tau_{\mathbf{k}xx}$ with coefficients depending on thermodynamic parameters, one may apply (A.2) to each of the terms separately. This yields

$$\begin{aligned} &(S_0, S_0) = k_{\mathrm{B}} C_P / n; \qquad (S_0, P_0) = (S_0, \tau_{0xx}) = 0 \\ &(P_0, P_0) = (P_0, \tau_{0xx}) = \rho c^2 / \beta \end{aligned} \tag{A.6}$$

In a similar way one can show that

$$(n_0, T_0) = 0; \qquad (T_0, T_0) = T / n\beta C_V \tag{A.7}$$

where $T_{\mathbf{k}}$ is defined in (129) and we introduced here the specific heats per particle C_P and C_V, and the adiabatic sound velocity c.

Next we consider the higher fluctuation formulas $(a_{\mathbf{k}}, b_{\mathbf{q}} c_{l})$ with $\mathbf{l} = \mathbf{k} - \mathbf{q}$, in the limit of small $\mathbf{k}$ and $\mathbf{q}$. We need only the following combinations, which have been evaluated by Ernst *et al.*[5]:

$$\begin{aligned} (\hat{\tau}_{0xx}, S_0 S_0) &= (n\beta)^{-2} \left\{ -\left(\frac{\partial P}{\partial e}\right)_n \left(\frac{\partial^2 e}{\partial T^2}\right)_P - \left(\frac{\partial P}{\partial n}\right)_e \left(\frac{\partial^2 n}{\partial T^2}\right)_P \right\} \\ &= \frac{k_{\mathrm{B}} C_P}{\beta n} (\gamma - 1) \left\{ 1 - \frac{1}{\alpha C_P} \left(\frac{\partial C_P}{\partial T}\right)_P + \frac{1}{\alpha^2} \left(\frac{\partial \alpha}{\partial T}\right)_P \right\} \end{aligned} \tag{A.8a}$$

$$(\hat{\tau}_{0xx}, S_0 P_0) = \left(\frac{n}{\beta}\right)^2 \left\{ \left(\frac{\partial S}{\partial \beta}\right)_v \left(\frac{\partial^2 \beta}{\partial n^2}\right)_S + \left(\frac{\partial S}{\partial v}\right)_\beta \left(\frac{\partial^2 v}{\partial n^2}\right)_S \right\} \tag{A.8b}$$

$$(\hat{\tau}_{0xx}, P_0 P_0) = \left(\frac{n}{\beta}\right)^2 \left\{ \left(\frac{\partial^2 P}{\partial n^2}\right)_S - \left(\frac{\partial P}{\partial e}\right)_n \left(\frac{\partial^2 e}{\partial n^2}\right)_S \right\}$$

$$= \frac{2\rho c^2}{\beta^2}\left\{\frac{n}{c}\left(\frac{\partial c}{\partial n}\right)_S - \frac{\gamma - 1}{2\alpha T}\right\} \tag{A.8c}$$

$$(\hat{\tau}_{0xx}, g_{0\alpha}g_{0\beta}) = \frac{2\rho}{\beta^2}\left\{\delta_{\alpha x}\delta_{\beta x} - \frac{\gamma - 1}{2\alpha T}\,\delta_{\alpha\beta}\right\} \tag{A.8d}$$

$$(\tau_{0xy}, g_{0\alpha}g_{0\beta}) = \frac{\rho}{\beta^2}\{\delta_{\alpha x}\delta_{\beta y} + \delta_{\alpha y}\delta_{\beta x}\} \tag{A.8e}$$

$$(\hat{j}_{0x}^{e}, g_{0x}S_0) = C_P/\beta^2 \tag{A.8f}$$

$$(\hat{j}_{0x}^{e}, g_{0x}P_0) = \rho c^2/\beta^2 \tag{A.8g}$$

where $\gamma = C_P/C_V$ and $\alpha = -n^{-1}(\partial n/\partial T)_P$. In (A.8d) and (A.8e) the labels α, β denote Cartesian components (x, y, z).

These fluctuation formulas are needed to evaluate the mode–mode amplitudes $A_i^{rs}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}})$ defined as

$$A_i^{rs}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = \langle \hat{j}_{kx}^{i}, a_{\mathbf{q}}{}^{r} a_{\boldsymbol{l}}{}^{s}\rangle \tag{A.9}$$

with $\boldsymbol{l} = \mathbf{k} - \mathbf{q}$, and $\hat{\mathbf{q}}$ and $\hat{\boldsymbol{l}}$ are unit vectors along $\mathbf{q}$ and $\boldsymbol{l}$, respectively. The label i takes the values η_i $(i = 1, 2)$, T, l, and σ $(\sigma = \pm 1)$, and the projected currents $\hat{j}_{kx}^{i}$ are defined in Eq. (42). The superscripts $(r, s) = (\eta_1, \eta_2, T, \sigma = \pm 1)$ label the hydrodynamic modes $a_{\mathbf{k}}{}^{i}$ defined in Eq. (17).

Using (A.8), we can immediately write down the expressions for all mode–mode amplitudes:

$$A_{\eta_1}^{\eta_i\eta_j}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\beta\rho)^{-1/2}\{\hat{q}_{\perp x}^{i}\hat{l}_{\perp y}^{j} + \hat{q}_{\perp y}^{i}\hat{l}_{\perp x}^{j}\} \tag{A.10}$$

$$A_{\eta_1}^{\sigma-\sigma}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = -(\beta\rho)^{-1/2}\tfrac{1}{2}\{\hat{q}_x\hat{l}_y + \hat{q}_y\hat{l}_x\} \tag{A.11}$$

$$A_T^{\eta_i T}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\beta\rho)^{-1/2}\hat{q}_{\perp x}^{i} \tag{A.12}$$

$$A_T^{\sigma-\sigma}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\beta\rho)^{-1/2}\tfrac{1}{2}\sigma[(\gamma - 1)^{1/2}/\alpha T](\hat{q}_x - \hat{l}_x) \tag{A.13}$$

$$A_l^{\eta_i\eta_j}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\beta\rho)^{-1/2}2\{\hat{q}_{\perp x}^{i}\hat{l}_{\perp x}^{j} - [(\gamma - 1)/2\alpha T]\hat{\mathbf{q}}_{\perp}{}^{i}\cdot\hat{\boldsymbol{l}}_{\perp}{}^{j}\} \tag{A.14}$$

$$A_l^{TT}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\beta\rho)^{-1/2}(\gamma - 1)\left\{1 - \frac{1}{\alpha C_P}\left(\frac{\partial C_P}{\partial T}\right)_P + \frac{1}{\alpha^2}\left(\frac{\partial \alpha}{\partial T}\right)_P\right\} \tag{A.15}$$

$$A_l^{\sigma-\sigma}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\beta\rho)^{-1/2}\left\{-\hat{q}_x\hat{l}_x - \frac{\gamma - 1}{2\alpha T}(1 - \hat{\mathbf{q}}\cdot\hat{\boldsymbol{l}}) + \frac{n}{c}\left(\frac{\partial c}{\partial n}\right)_S\right\} \tag{A.16}$$

$$A_{\sigma'}^{\eta_i\eta_j}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\sigma'/\sqrt{2})A_l^{\eta_i\eta_j}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) \tag{A.17}$$

$$A_{\sigma'}^{\eta_i T}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = [(\gamma - 1)^{1/2}/\sqrt{2}]A_T^{\eta_i T}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) \tag{A.18}$$

$$A_{\sigma'}^{TT}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = (\sigma'/\sqrt{2})A_l^{TT}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) \tag{A.19}$$

$$A_{\sigma'}^{\sigma-\sigma}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) = \frac{(\gamma - 1)^{1/2}}{\sqrt{2}}A_T^{\sigma-\sigma}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) + \frac{\sigma'}{\sqrt{2}}A_l^{\sigma-\sigma}(\hat{\mathbf{q}}, \hat{\boldsymbol{l}}) \tag{A.20}$$

The set of unit vectors $\hat{\mathbf{q}}$, $\hat{\mathbf{q}}_\perp^i$ ($i = 1, 2$) are mutually orthogonal, as are the set $\hat{l}$, $\hat{l}_\perp^i$ ($i = 1, 2$). The amplitudes with $(r, s) = (\sigma\sigma)$, (σT), and $(\sigma\eta_i)$ are not listed, since they are not needed in our theory. All other unlisted amplitudes vanish.

In order to have a complete list of relevant amplitudes, it is worthwhile to consider the amplitudes involved in the correlation function $C_{\zeta\zeta}(0, z)$ for the bulk viscosity, which are defined as

$$A_\zeta^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta/\rho)^{1/2}\tfrac{1}{3} \sum_{x=x,y,z} (\hat{\tau}_{0xx}, a_{\mathbf{q}}{}^a a^b_{-\mathbf{q}}) = \tfrac{1}{3} \sum_{x=x,y,z} A_l^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \quad \text{(A.21)}$$

and we find from (A.14)–(A.16)

$$A_\zeta^{\eta_i\eta_j}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = -(\beta\rho)^{-1/2}\left\{\frac{2}{3} - \frac{\gamma - 1}{\alpha T}\right\}\delta_{ij} \qquad (i,j = 1, 2) \qquad \text{(A.22)}$$

$$A_\zeta^{TT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1/2}(\gamma - 1)\left\{1 - \frac{1}{\alpha C_P}\left(\frac{\partial C_P}{\partial T}\right)_P + \frac{1}{\alpha^2}\left(\frac{\partial \alpha}{\partial T}\right)_P\right\} \quad \text{(A.23)}$$

$$A_\zeta^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1/2}\left\{\frac{1}{3} - \frac{\gamma - 1}{\alpha T} + \frac{n}{c}\left(\frac{\partial c}{\partial n}\right)_s\right\} \qquad \text{(A.24)}$$

APPENDIX B. THE MODE–MODE INTEGRALS FOR SMALL VALUES OF k AND z

The mode–mode integrals have the general form

$$U_{ij}^{ab}(\mathbf{k}, z) = \int' \frac{d\mathbf{q}}{(2\pi)^3} \frac{S_{ij}^{ab}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)}{z - z_a(q_1) - z_b(q_2)} \qquad \text{(B.1)}$$

where $\mathbf{q}_1 + \mathbf{q}_2 = \mathbf{k}$, and we take $\mathbf{k} = \mathbf{k}\hat{\mathbf{x}}$. It will be convenient to write $\mathbf{q}_1 = \mathbf{q} + \gamma_1\mathbf{k}$ and $\mathbf{q}_2 = -\mathbf{q} + \gamma_2\mathbf{k}$, where $\gamma_1 = 1 - \gamma_2$ is an arbitrary real number. This is done in order to simplify a number of integrals which will appear later. The prime on the integral sign indicates that $|\mathbf{q}| < k_0$. The vectors $\hat{\mathbf{q}}_1$ and $\hat{\mathbf{q}}_2$ are unit vectors parallel to $\mathbf{q}_1$ and $\mathbf{q}_2$, respectively.

We make the following ansatz for the frequencies $z_a(k)$ for small k with $\alpha = \sigma, \eta, T$:

$$\begin{aligned} z_\mu(k) &= -D_\mu k^2 + \Delta_\mu k^{2+P}, & \mu &= \eta, T \\ z_\sigma(k) &= -i\sigma ck - D_s k^2 + (\Delta_s - i\sigma\overline{\Delta}_s)k^{2+P}, & \sigma &= \pm \end{aligned} \qquad \text{(B.2)}$$

where $\frac{1}{2} \leqslant P < 1$. The coefficient of k^{2+P} in z_σ is in general complex. From the observation $z_\sigma^* = z_{-\sigma}$, made in Section 3, it follows that the real part of this coefficient is an even function of σ, and its imaginary part is an odd function of σ.

In the following discussion we investigate the behavior of $U_{ij}^{ab}(\mathbf{k}, z)$ for small values of k and z, starting with $U_{ij}^{\sigma-\sigma}(\mathbf{k}, z)$, For this case we choose

$\gamma_1 = \gamma_2 = \frac{1}{2}$, so that $q_1{}^2 + q_2{}^2$ in the denominator of (B.1) equals $2q^2 + \frac{1}{2}k^2$. The dominant contributions to $U_{ij}^{\sigma-\sigma}(\mathbf{k}, z)$ come from regions where the denominator of (B.1) becomes small, i.e., from regions around the lower limit of the q-integration. In this region the denominator approaches $z + i\sigma ckx$, where $x = \hat{\mathbf{k}}\cdot\hat{\mathbf{q}}$, and the larger of the two small parameters z and k will determine the dominant behavior of the integral. Therefore we introduce a small parameter $\theta = \max(z, k)$, which is equal to the maximum of z and k. It is convenient to divide the interval for the q-integration into two regions: $0 < q < N\theta$ $(N > 1)$ and $N\theta < q < k_0$. In the first region we make the substitution $q = \theta y$, and we determine the leading term for small θ. The function $S^{\sigma-\sigma}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)$ can be bounded from above by a constant; the denominator approaches $\{z + i\sigma c(y_1 - y_2) + O(\theta^2)\}$, where $y_i = \theta^{-1}q_i$. Therefore the contribution from the first region to $U^{\sigma-\sigma}(\mathbf{k}, z)$ is of $O(\theta^2)$, and we can write (B.1) as

$$U^{\sigma-\sigma}(\mathbf{k}, z) = \int \frac{d\hat{\mathbf{q}}}{(2\pi)^3} \int_{N\theta}^{k_0} dq\, q^2 \frac{S^{\sigma-\sigma}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)}{-z_\sigma(q) - z_{-\sigma}(q)} \{1 - F(q)\} + O(\theta^2) \tag{B.3}$$

where

$$F(q) = \frac{z - z_\sigma(q_1) - z_{-\sigma}(q_2) + z_\sigma(q) + z_{-\sigma}(q)}{z - z_\sigma(q_1) - z_{-\sigma}(q_2)} \tag{B.4}$$

The q values in (B.3) are always larger than k, so that we may expand the integrand of (B.3) in powers of k. Consider first the functions $S_{ij}^{\sigma-\sigma}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)$, which, according to (66), are given in terms of the amplitudes $A_i^{\sigma-\sigma}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)$. The amplitudes, calculated in (A.10)–(A.24), depend on $\hat{\mathbf{q}}_1$ and $\hat{\mathbf{q}}_2$ only through the combinations $\hat{q}_{1\alpha}\hat{q}_{2\alpha}$ $(\alpha = x, y, z)$, $(\hat{q}_{1x}\hat{q}_{2y} + \hat{q}_{1y}\hat{q}_{2x})$, and $(\hat{q}_{1x} - \hat{q}_{2x})$. These functions are obviously even functions of k. For small values of k these combinations are equal to their value at $k = 0$, plus a correction term of relative order $O(k^2q^{-2})$. Hence,

$$S^{\sigma-\sigma}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2) = S^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})[1 + O(k^2q^{-2})]$$

The integrand in (B.3) consists of two terms; one term, which does not contain $F(q)$, yields a constant $U^{\sigma-\sigma}(0, 0)$, plus a correction term of $O(\theta)$ from the lower limit, plus a correction term of $O(k^2\theta^{-1}) \leqslant O(\theta)$, which results from the $O(k^2q^{-2})$ correction to $S^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$. In the second term of (B.3), containing $F(q)$, we make the substitution $q = \theta^{1/2}y$, so that (B.3) can be written as

$$\begin{aligned} U^{\sigma-\sigma}(\mathbf{k}, z) = {} & U^{\sigma-\sigma}(0, 0) \\ & - \frac{\theta^{1/2}}{(2\pi)^3} \int d\hat{\mathbf{q}} \int_{N\sqrt{\theta}}^{k_0/\sqrt{\theta}} dy\, \frac{S^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})[1 + O(k^2y^{-2}\theta^{-1})}{2D_s - 2\Delta_s y^P \theta^{P/2}} \\ & \times F(\theta^{1/2}y) + O(\theta) \end{aligned} \tag{B.5}$$

where

$$F(\theta^{1/2}y) = \frac{z + i\sigma ckx + O(k^3y^{-2}\theta^{-1}) + O(ky^{1+P}\theta^{\frac{1}{2}+\frac{1}{2}P})}{z + i\sigma ckx + 2D_s y^2\theta - 2\Delta_s y^{2+P}\theta^{1+\frac{1}{2}P} + O(k^3y^{-2}\theta^{-1}) + O(ky^{1+P}\theta^{\frac{1}{2}+\frac{1}{2}P})} \tag{B.6}$$

One can easily estimate the correction terms in (B.5) and (B.6) to be at least of $O(\theta)$.

The numerator and denominator in the integrand of (B.5) still contain θ-dependent terms of relative order $\theta^{P/2}$. The coefficient of the first correction term, which is of order $\theta^{\frac{1}{2}+\frac{1}{2}P}$, can be determined from (B.5) by expanding the integrand in powers of $\theta^{P/2}$, and keeping only terms linear in $\theta^{P/2}$. The result of (B.5), correct to $O(\theta)$, is then given by

$$U^{\sigma-\sigma}(\mathbf{k}, z) = U^{\sigma-\sigma}(0, 0) - \int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{4\pi^2 D_s} \int_0^\infty dq \frac{\zeta}{\zeta + q^2}\left[1 + q^P \frac{\Delta_s}{D_s}\frac{\zeta + 2q^2}{\zeta + q^2}\right] + O(\theta) \tag{B.7}$$

where we have extended the y-integration in (B.5) to the interval $(0, \infty)$, which again introduces errors of $O(\theta)$. In (B.7) we have changed back to the original integration variables and we introduced

$$\zeta = (z + i\sigma ckx)/2D_s \tag{B.8}$$

This result implies also that z in (B.8) should be replaced by zero if $z = O(k^2)$.

As the next step we evaluate the q-integral in (B.7) for real, positive ζ, and extend the result to complex ζ by analytic continuation. Since the integral

$$\int_0^\infty dq\, q^P \frac{\zeta(\zeta + 2q^2)}{(\zeta + q^2)^2} = \zeta^{(P+1)/2}\frac{\pi(P + 3)}{4\cos\frac{1}{2}\pi P} \tag{B.9}$$

we find from (B.7)

$$U^{\sigma-\sigma}(\mathbf{k}, z) = U^{\sigma-\sigma}(0, 0) - \int \frac{d\hat{\mathbf{q}}}{4\pi}\frac{S^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{8\pi D_s}\left[\zeta^{1/2} + \frac{\Delta_s}{D_s}\frac{P + 3}{2\cos\frac{1}{2}\pi P}\zeta^{(P+1)/2}\right] + O(\theta) \tag{B.10}$$

where broken powers of ζ are uniquely defined for complex ζ by introducing a branch cut along the negative real axis from $\zeta = 0$ toward minus infinity.

For a discussion of $U_{ij}^{\lambda\mu}(\mathbf{k}, z)$, where (λ, μ) label diffusive modes (η_1, η_2, T), we choose $\gamma_1 = 1 - \gamma_2 = D_\mu/(2D_{\lambda\mu})$, with $2D_{\lambda\mu} = D_\lambda + D_\mu$, so that

$$D_\lambda {q_1}^2 + D_\mu {q_2}^2 = 2D_{\lambda\mu}q^2 + k^2(D_\lambda D_\mu/2D_{\lambda\mu})$$

We divide the q-integration again into the regions $0 < q < N\theta$ and $N\theta < q < k_0$ with $N > 1$ and $\theta = \max(z, k)$. The contribution from the first region is estimated in the same way as before, and yields a term of $O(\theta^3\zeta^{-1})$, where $\zeta = (2D_{\lambda\mu})^{-1}[z + (D_\lambda D_\mu/2D_{\lambda\mu})k^2]$. We then arrive at an expression similar to (B.3) and (B.4), in which σ and $-\sigma$ are replaced by λ and μ, respectively, and the term of $O(\theta^2)$ in (B.3) is replaced by $O(\theta^3\zeta^{-1}) \leqslant O(\theta)$. In order to proceed, we have to consider first the k-expansion of the functions $S^{\lambda\mu}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)$. In the previous case the strength factor $S^{\sigma-\sigma}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)$ contained only even powers of k. This property remains valid whenever two heat modes or two shear modes are involved. However, for strength factors $S^{\eta_i T}(\hat{\mathbf{q}}_1, \hat{\mathbf{q}}_2)$ containing a heat mode and a shear mode, odd powers of k also enter into the k-expansion of the strength factors. The terms involving odd powers of k have coefficients that are odd functions of x, while terms involving even powers of k have coefficients even in x. From here on the analysis proceeds in the same way as before, using in addition that odd functions of x vanish due to angular q-integrations. The final result then reads

$$U^{\lambda\mu}(\mathbf{k}, z) = U^{\lambda\mu}(0, 0) - \int \frac{d\hat{\mathbf{q}}}{4\pi} \frac{S^{\lambda\mu}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})}{8\pi D_{\lambda\mu}} \left[\zeta^{1/2} + \frac{\Delta_{\lambda\mu}(P+3)}{D_{\lambda\mu} 2\cos\frac{1}{2}\pi P} \zeta^{(P+1)/2}\right] + O(\theta) \quad \text{(B.11)}$$

The implications of this result are as follows. If $z \leqslant O(k^2)$, so that $\theta = k$ and $\zeta = O(k^2)$, then the leading term in (B.11) is proportional to $\sqrt{\zeta} = O(k)$, and is of the same order as the neglected terms of $O(\theta)$. The only conclusion to be drawn is then

$$U^{\lambda\mu}(\mathbf{k}, z) = U^{\lambda\mu}(0, 0) + O(k) \qquad \text{for} \quad z \leqslant O(k^2) \quad \text{(B.12)}$$

The term in (B.11) proportional to $\zeta^{(P+1)/2}$ is only meaningful when it is larger than $O(\theta)$, which is the case for $z > O(k^{2/(P+1)})$. In Section 3 we use (B.11) for P in the range $\frac{1}{2} \leqslant P < 1$; so we should restrict z to be larger than or equal to $O(k)$, and we should therefore neglect the terms of $O(k^2)$ in ζ. Thus Eq. (B.11) holds if

$$z \geqslant O(k) \quad \text{(B.13)}$$

and

$$\zeta = z/2D_{\lambda\mu} \quad \text{(B.14)}$$

The remaining mode–mode integrals $U_{ij}^{\sigma\sigma}(\mathbf{k}, z)$ and $U_{ij}^{ab}(\mathbf{k}, z)$ can be estimated with the result

$$U_{ij}^{\sigma\sigma}(\mathbf{k}, z) = U_{ij}^{\sigma\sigma}(0, 0) + O(\theta) \quad \text{(B.15)}$$

and a similar result for $U^{\sigma\lambda}(\mathbf{k}, z)$.

An inspection of three-mode contributions to $U_{ij}^{ab}(\mathbf{k}, z)$ and of contributions from the higher terms in the k-expansion of the hydrodynamic modes indicates that these contributions are less important than the ones considered here.

APPENDIX C

In this appendix the mode–mode strength factors $S_{ij}^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})$ are calculated, as well as the coefficients $M_{ij}^{ab}(n)$, which involve certain angular averages of the strength factors.

From the results of Appendix A the strength factors, for $k = 0$, defined as

$$S_{ij}^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = A_i^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})(A_j^{ab}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}))^* \tag{C.1}$$

can be calculated in a straightforward manner. It is convenient to take all degenerate shear mode contributions together, using the identity

$$\hat{q}_\alpha \hat{q}_\beta + \sum_{i=1,2} \hat{q}_{\perp\alpha}^i \hat{q}_{\perp\beta}^i = \delta_{\alpha\beta} \tag{C.2}$$

where $\alpha, \beta = x, y, z$ label Cartesian components. The results are

$$\sum_{i,j=1}^{2} S_{\eta_1\eta_1}^{\eta_i\eta_j}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}2[1 - \hat{q}_x^2 - \hat{q}_y^2 + 2\hat{q}_x^2\hat{q}_y^2] \tag{C.3a}$$

$$= (\beta\rho)^{-1}(1 + x^2 - 2x^4) \tag{C.3b}$$

$$S_{\eta_1\eta_1}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}\hat{q}_x^2\hat{q}_y^2 \tag{C.4a}$$

$$= (2\beta\rho)^{-1}x^2(1 - x^2) \tag{C.4b}$$

$$\sum_{i=1}^{2} S_{TT}^{\eta_i T}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}(1 - x^2) \tag{C.5}$$

$$S_{TT}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}(\gamma - 1)(\alpha T)^{-2}x^2 \tag{C.6}$$

$$\sum_{i,j=1}^{2} S_{ll}^{\eta_i\eta_j}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}4[(1 - x^2)^2 + A(1 - x^2) + \tfrac{1}{2}A^2] \tag{C.7}$$

$$S_{ll}^{TT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}B^2 \tag{C.8}$$

$$S_{ll}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}[x^2 + A + D]^2 \tag{C.9}$$

$$S_{lT}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}\frac{(\gamma - 1)^{1/2}}{\alpha T}\sigma x[x^2 + A + D] \tag{C.10}$$

$$\sum_{i,j=1}^{2} S_{\sigma\sigma}^{\eta_i\eta_j}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}2[(1 - x^2)^2 + A(1 - x^2) + \tfrac{1}{2}A^2] \tag{C.11}$$

$$\sum_{i=1}^{2} S_{\sigma\sigma}^{\eta_i T}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}(\gamma - 1)\tfrac{1}{2}(1 - x^2) \tag{C.12}$$

$$S_{\sigma\sigma}^{TT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}\tfrac{1}{2}B^2 \tag{C.13}$$

$$S_{\sigma\sigma}^{\sigma'-\sigma'}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}\tfrac{1}{2}[x^2 + A(1 - \sigma\sigma' x) + D]^2 \tag{C.14a}$$

$$\equiv (\beta\rho)^{-1}M(\sigma\sigma' x) \tag{C.14b}$$

$$\sum_{i,j=1}^{2} S_{\zeta\zeta}^{\eta_i\eta_j}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}2[\tfrac{2}{3} + A]^2 \tag{C.15}$$

$$S_{\zeta\zeta}^{TT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}B^2 \tag{C.16}$$

$$S_{\zeta\zeta}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) = (\beta\rho)^{-1}[\tfrac{1}{3} + A + D]^2 \tag{C.17}$$

where

$$A = -\frac{\gamma - 1}{\alpha T}; \qquad D = \frac{n}{c}\left(\frac{\partial c}{\partial n}\right)_S$$
$$B = \left[1 - \frac{1}{\alpha C_P}\left(\frac{\partial C_P}{\partial T}\right)_P + \frac{1}{\alpha^2}\left(\frac{\partial \alpha}{\partial T}\right)_P\right](\gamma - 1) \tag{C.18}$$

In these expressions $x = \hat{q}_x$. We notice further that the equalities (C.3b) and (C.4b) are actually the averages of (C.3a) and (C.4a), respectively, over the azimuthal angle. The equality sign applies only under the integral sign $\int d\hat{\mathbf{q}}$, which is all that is needed.

In the second part of this appendix the quantities $M_{ii}^{ab}(n)$ defined in (76) are calculated, starting with ($\mu = \eta, T$)

$$M_{\mu\mu}^{ss}(n) = \frac{1}{2}\beta\rho \sum_{\sigma} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{\mu\mu}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})|x|^{P_n} \tag{C.19}$$

Using (C.4) and (C.6), this yields

$$M_{\eta\eta}^{ss}(n) = [(3 + P_n)(5 + P_n)]^{-1} \tag{C.20}$$

$$M_{TT}^{ss}(n) = (\gamma - 1)(\alpha T)^{-2}(3 + P_n)^{-1} \tag{C.21}$$

The quantity $M_{\sigma\sigma}^{ss}(n)$ is defined, according to (85b), as

$$M_{\sigma\sigma}^{ss}(n) = \tfrac{1}{2}\int_{-1}^{1} dx\, M(x)(1 - x)^{P_n} \tag{C.22}$$

where $P_n = 1 - 2^{-n}$. From (C.14) we find

$$M_{\sigma\sigma}^{ss}(0) = \tfrac{1}{2}(\tfrac{1}{5} + \tfrac{2}{3}(A + D) + \tfrac{4}{3}A^2 + 2AD + D^2) \tag{C.23}$$

$$M_{\sigma\sigma}^{ss}(1) = \sqrt{2}\left(\frac{211}{5\cdot7\cdot9\cdot11} + \frac{92}{5\cdot7\cdot9}A + \frac{22}{3\cdot5\cdot7}D + \frac{4}{7}A^2 + \frac{4}{5}AD + \frac{1}{3}D^2\right) \tag{C.24}$$

$$\vdots$$

$$M_{\sigma\sigma}^{ss}(\infty) = \tfrac{1}{2}(\tfrac{1}{5} + \tfrac{16}{15}A + \tfrac{2}{3}D + 2A^2 + \tfrac{8}{3}AD + D^2) \tag{C.25}$$

Table I. Values of $M_{rr}^{ab}(0)$

$rr \backslash ab$	ss	$\eta\eta$	ηT	TT
$\eta_1\eta_1$	$\frac{1}{15}$	$\frac{7}{15}$	0	0
TT	$\frac{1}{3}(\gamma - 1)/(\alpha T)^2$	0	$\frac{2}{3}$	0
ll	$\frac{1}{5} + \frac{2}{3}(A + D) + (A + D)^2$	$\frac{16}{15} + \frac{4}{3}A + A^2$	0	$\frac{1}{2}B^2$
$\zeta\zeta$	$(\frac{1}{3} + A + D)^2$	$(\frac{2}{3} + A)^2$	0	$\frac{1}{2}B^2$
$\sigma\sigma$	$\frac{1}{2}[\frac{1}{5} + \frac{2}{3}(A + D) + \frac{4}{3}A^2 + 2AD + D^2]$	$\frac{8}{15} + \frac{2}{3}A + \frac{1}{2}A^2$	$\frac{1}{3}(\gamma - 1)$	$\frac{1}{4}B^2$

The next set of quantities to be considered are $M_{rr}^{ss}(0)$, $M_{rr}^{\eta\eta}(0)$, $M_{rr}^{\eta T}(0)$, and $M_{rr}^{TT}(0)$ for $r = \sigma, \eta_1, T, l$, and ζ, defined as

$$
\begin{aligned}
M_{rr}^{ss}(0) &= \frac{1}{2}\beta\rho \sum_{\sigma} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{rr}^{\sigma-\sigma}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \\
M_{rr}^{\eta\eta}(0) &= \frac{1}{2}\beta\rho \sum_{i,j} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{rr}^{\eta_i\eta_j}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \\
M_{rr}^{\eta T}(0) &= \beta\rho \sum_{i} \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{rr}^{\eta_i T}(\hat{\mathbf{q}}, -\hat{\mathbf{q}}) \\
M_{rr}^{TT}(0) &= \frac{1}{2}\beta\rho \int \frac{d\hat{\mathbf{q}}}{4\pi} S_{rr}^{TT}(\hat{\mathbf{q}}, -\hat{\mathbf{q}})
\end{aligned}
\tag{C.26}
$$

Notice that (C.19) and (C.22) reduce to (C.26) for $n = 0$. The results are given in Table I.

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