PHYSICAL REVIEW A VOLUME 32, NUMBER 5 NOVEMBER 1985

Hydrodynamic fluctuations at large shear rate

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(Received 14 May 1985)

The nonlinear Navier-Stokes-Langevin equations are used to describe fluctuations in a compressible fluid with uniform shear flow. The hydrodynamic modes for small deviations from the macroscopic nonequilibrium state are calculated, including linear mode coupling of the fluctuating variables with the macroscopic velocity field. The associated correlation functions are determined with the full nonlinear dependence on shear rate required for long times and/or large shear rate. The stationary and joint probability densities are also constructed from the associated Fokker-Planck equation. As an application of these results, the lowest-order mode-coupling contributions to the renormalized shear viscosity are evaluated.

I. INTRODUCTION

A simple fluid in uniform shear flow is characterized by a velocity field with constant gradient orthogonal to the flow. The magnitude of the gradient, or shear rate, provides a single control parameter to measure the departure of the fluid from its equilibrium state. For planar geometry, the pressure is spatially constant and the temperature is either temporally or spatially constant (depending on boundary conditions), so the macroscopic state of the fluid can be very far from equilibrium, for large shear route, but still structurally quite simple. Because of this relative simplicity, such a system provides a convenient testing ground for the concepts and methods of nonequilibrium statistical mechanics.[1] In recent years there have been several attempts to calculate the transport and fluctuation properties for shear flow, both theoretically[2–12] and by novel methods of nonequilibrium computer simulation;[13–19] some experiments also have been performed.[20–23] Many of the most interesting features observed in these results are attributed to the coupling of hydrodynamic modes in the nonequilibrium fluid. The objective here is to describe in some detail these hydrodynamic modes for a compressible fluid up to quite large shear rates. The results allow calculation of the hydrodynamic fluctuations, or time correlation functions, and related transport properties for this nonequilibrium state. In the limit of small shear rate, the results obtained here reduce to previous calculations generally limited to first order in the shear rate, and at zero shear rate they reduce to the well-known expressions for hydrodynamic fluctuations in an equilibrium fluid.[24]

There are several motivations for continued attention to the properties of a fluid under shear. The nonequilibrium computer simulations mentioned above have provided a great deal of data that do not have an adequate theoretical explanation. For example, the shear viscosity is expected to decrease with increasing shear rate (shear thinning). At small shear rates a, the hydrodynamic mode-coupling theories predict that the shear viscosity in three dimensions has the form[2]

$$\eta(a)\sim\eta_0+\eta_1 a^{1/2},\ \ \eta_1<0\ . \tag{1.1}$$

The surprising feature of this result is the nonanalytic dependence on the shear rate, indicating a divergence of formal nonlinear response functions. While the computer-simulation results can be fit to this $a^{1/2}$ dependence, the coefficient η_1 obtained in this way differs by two orders of magnitude from the theoretical values.[14] Although there has been a recent suggestion for the possible resolution of this discrepancy,[25] the problem remains open. In two dimensions the mode-coupling theory predicts a logarithmic nonanalyticity, and at sufficiently large shear rate the computer-simulation data can be fit to this functional form but with a coefficient that is again too large as compared to theory. Furthermore, at smaller shear rates in two dimensions, computer-simulation results suggest that the shear viscosity is independent of shear rate. One explanation for this[14] is a small-shear-rate instability that is not accounted for in the theoretical models. Finally, and perhaps most puzzling, is a structural phase transition observed recently in a three-dimensional computer simulation at large shear rate.[19] At a critical shear rate enhanced shear thinning was observed, while above this shear rate the particles (hard spheres) were found to be localized in tubes along the direction of flow, with a hexagonal packing of the tubes. Since planar Couette flow is expected to be linearly stable[26] (as confirmed here), the mechanism for such a transition is not evident. A related class of mode-coupling problems for the hydrodynamics of liquid crystals under shear has been discussed also.[27]

The discussion given here does not address these questions directly. However, it is generally assumed that all of the above phenomena are related to the nonlinear dynamics of hydrodynamic fluctuations, which describe the coupling of the thermal fluctuations around the nonequilibrium macroscopic state. From the corresponding study of nonlinear mode coupling of excitations around equilibrium, it may be expected that such effects are singular in two dimensions,[28] but can be treated perturbatively in three dimensions. Much less is known about such mode coupling for a nonequilibrium state (particularly for a compressible fluid), and one objective here is to provide the nonequilibrium hydrodynamic modes required for such a mode-coupling analysis. To illustrate their utility in this context, the fluctuation-renormalized shear viscosity is calculated and the coefficient η_1 in Eq. (1.1) is determined.[29]

The model used to describe hydrodynamic fluctuations is the Navier-Stokes-Langevin equations. This is a set of five coupled equations for the local conserved densities (mass, energy, and momentum) that is structurally the same as the macroscopic nonlinear Navier-Stokes equations. However, the heat flux and pressure tensor have fluctuating components that represent the dynamics of all other (presumably more rapidly varying) degrees of freedom. These fluctuating components are then modeled by a Gaussian-Markovian process. The consistency of such nonlinear Langevin equations with basic principles of irreversible thermodynamics has been doubted,[30] but recent results have largely dispelled such questions.[31] The precise definition of the Navier-Stokes-Langevin model is given in Sec. II and Appendix A. These equations are then linearized around the state of uniform shear flow and diagonalized. The resulting set of five independent linear equations defines the hydrodynamic modes. The calculation parallels closely that for the equilibrium state.[32] In the latter case the modes are parametrized by a single wave vector $\mathbf{k}$, while in the nonequilibrium case there is a coupling of these wave vectors even in the linearized equation. This linear mode coupling is due to terms that are bilinear in the fluctuations and the macroscopic flow velocity.[33] The latter behaves as an external inhomogeneous field that induces the wave-vector coupling, leading to qualitative differences from the equilibrium hydrodynamic modes (e.g., nonexponential time dependence), and is essential for the stability of these modes at large shear rate. The equations are solved in a manner valid for large shear rates in the sense that secular terms $\sim(at)$ are included to all orders. It is still possible to interpret the results as two sound modes, a heat mode, and two shear modes, although the equilibrium degeneracy of the latter is broken in the nonequilibrium state.

The matrix of time correlation functions is calculated from these modes in Sec. IV. These functions obey a generalization of Onsager's assumption[34] on the regression of fluctuations to nonequilibrium states.[35] The equal-time correlation functions are shown to exhibit long-range order with a correlation length $l\sim(\Gamma/a)^{1/2}$, where Γ is the sound damping constant. The dynamic structure factor for the Brillouin light scattering spectrum is identified as the sum of Brillouin and Rayleigh peaks which, in contrast to equilibrium fluids, are not simply Lorentzian shapes. The Landau-Placzek ratio of integrated intensities is calculated and found to be a function of both wave vector and shear rate. In Sec. V the Fokker-Planck equation associated with the linearized Navier-Stokes-Langevin equation is constructed, and the stationary state and the joint probability densities are determined. Finally, the results are summarized and discussed in Sec. VI.

II. NAVIER-STOKES-LANGEVIN EQUATIONS

The microscopic conserved densities for a simple fluid are linear combinations of the mass density $\rho(\mathbf{r},t)$, the energy density $u(\mathbf{r},t)$, and the momentum density $\mathbf{p}(\mathbf{r},t)$. The local conservation laws are given by

$$
\begin{aligned}
&\frac{\partial}{\partial t}\rho(\mathbf{r},t)+\nabla\cdot\mathbf{p}(\mathbf{r},t)=0\ ,\\
&\frac{\partial}{\partial t}u(\mathbf{r},t)+\nabla\cdot\mathbf{s}(\mathbf{r},t)=0\ ,\\
&\frac{\partial}{\partial t}p_i(\mathbf{r},t)+\frac{\partial}{\partial r_j}t_{ij}(\mathbf{r},t)=0\ ,
\end{aligned}
\tag{2.1}
$$

where $\mathbf{s}(\mathbf{r},t)$ and $t_{ij}(\mathbf{r},t)$ are the associated energy and momentum fluxes. The specific forms of these phase functions are well known[36] and will not be given here. By analogy with the macroscopic hydrodynamic equations it is convenient to express the energy density and fluxes in terms of the corresponding quantities referred to a frame of reference locally at rest with respect to the fluid. This can be accomplished in a formal way by introducing a local microscopic velocity field by $\mathbf{p}(\mathbf{r},t)=\rho(\mathbf{r},t)\mathbf{v}(\mathbf{r},t)$. Then a Galilean transformation of the densities and fluxes leads to

$$
\begin{aligned}
&\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla\right]\rho+\rho\nabla\cdot\mathbf{v}=0\ ,\\
&\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla\right]u'+u'\nabla\cdot\mathbf{v}+\nabla\cdot\mathbf{s}'+t'_{ij}\frac{\partial v_i}{\partial r_j}=0\ ,\\
&\rho\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla\right]v_i+\frac{\partial}{\partial r_j}t'_{ij}=0\ ,
\end{aligned}
\tag{2.2}
$$

where the quantities with a prime denote the corresponding phase function referred to the local rest frame. In this form Eqs. (2.2) resemble closely the macroscopic conservation laws. To complete the parallel, the rest-frame fluxes are further decomposed into two parts, one whose dependence on the microscopic degrees of freedom occurs only through the conserved densities ρ and u' and a remaining "random component" which cannot be written in that form

$$
\begin{aligned}
&\mathbf{s}'=\mathbf{s}^*(\rho,u')+\mathbf{s}^R\ ,\\
&t'_{ij}=p(\rho,u')\delta_{ij}+t^*_{ij}(\rho,u')+t^R_{ij}\ .
\end{aligned}
\tag{2.3}
$$

In the second equation of (2.3) a form analogous to that for the macroscopic equations has been chosen, where the microscopic pressure has been introduced. It is defined to be the same function of ρ and u' as in the equilibrium equation of state,

$$
p(\rho,u')\equiv p_e(\rho,u')\ . \tag{2.4}
$$

Equations (2.2) and (2.3) are still exact but not very useful until the functional forms of s^* and t^* are specified. A model Langevin equation results from idealizing the detailed structure of the fast degrees of freedom as a Gaussian-Markovian process.[30,31] This is accomplished by taking the random components as linear functionals of the corresponding stochastic variables. Considerations of irreversible thermodynamics then impose relationships[31] between the covariances of these variables and the forms of s_i^* and t_{ij}^*. For processes with small spatial variations, the latter are assumed to have the Navier-Stokes form

$$\mathbf{s}^* = -\lambda_B \nabla T \ ,$$
$$t_{ij}^* = -\eta_B \left[\frac{\partial v_i}{\partial r_j} + \frac{\partial v_j}{\partial r_i} - \frac{2}{3}\delta_{ij}\nabla\cdot\mathbf{v} \right] - \kappa_B \nabla\cdot\mathbf{v}\delta_{ij} \ . \tag{2.5}$$

Here λ_B, η_B, and κ_B are "bare" transport coefficients that depend in general on ρ and u'. Also, the microscopic temperature T is defined in a manner similar to (2.4),

$$T = T_e(\rho, u') \ , \tag{2.6}$$

where T_e is the equilibrium function of its arguments. Equations (2.2)–(2.6) together with the Gaussian-Markovian properties of the random components give a closed set of five nonlinear stochastic differential equations, the Navier-Stokes-Langevin equations. A more precise specification is given in Appendix A. The physical basis for the idealization involved in such Langevin models has been discussed at length elsewhere[30,31] and no further comment will be made here.

For notational simplicity a five-dimensional vector whose components are the conserved densities is introduced:

$$y \leftrightarrow (\rho(\mathbf{r},t),\, u(\mathbf{r},t),\, p_i(\mathbf{r},t)) \ . \tag{2.7}$$

The vector whose components are the *nonequilibrium* averages of these densities will be denoted by y_0,

$$y_0 \leftrightarrow (\rho_0(\mathbf{r},t),\, u_0(\mathbf{r},t),\, p_{0i}(\mathbf{r},t)) \ , \tag{2.8}$$

where a subscript, 0, will be used to indicate an averaged quantity. To study small fluctuations around a given macroscopic state, the relevant variables are the deviations of the conserved densities from their average values,

$$z(\mathbf{r},t) \equiv y(\mathbf{r},t) - y_0(\mathbf{r},t) \ . \tag{2.9}$$

Linearization of the Navier-Stokes-Langevin equations with respect to $z(\mathbf{r},t)$ leads to the general form

$$\frac{\partial}{\partial t} z_\alpha(\mathbf{r},t) + \mathscr{L}_{\alpha\beta}(y_0(\mathbf{r},t)) z_\beta(\mathbf{r},t) = R_\alpha(\mathbf{r},t) \ , \tag{2.10}$$

where $R_\alpha(\mathbf{r},t)$ contains all of the contribution from the random components in (2.3), and the dependence of the linear operator $\mathscr{L}_{\alpha\beta}$ on the particular macroscopic state considered has been made explicit. In terms of the Fourier transform of $z(\mathbf{r},t)$ these equations have the form

$$\frac{\partial}{\partial t}\tilde{z}_\alpha(\mathbf{k},t) + \int d\mathbf{k}' \tilde{\mathscr{L}}_{\alpha\beta}(\mathbf{k},\mathbf{k}';t)\tilde{z}_\beta(\mathbf{k}',t) = \tilde{R}_\alpha(\mathbf{k},t) \ , \tag{2.11}$$

where

$$\tilde{z}(\mathbf{k},t) \equiv \int d\mathbf{r}\, e^{i\mathbf{k}\cdot\mathbf{r}} z(\mathbf{r},t) \tag{2.12}$$

and $\tilde{R}_\alpha$ is the transform of R_α. The solutions to these linear Langevin equations define the hydrodynamic modes. The nonlocal form of the matrix, $\tilde{\mathscr{L}}(\mathbf{k},\mathbf{k};t)$, with respect to $\mathbf{k}$ is a result of the inhomogeneities of the macroscopic variables $y_0(\mathbf{r},t)$ in Eq. (2.10). For the equilibrium state $\mathscr{L}_{\alpha\beta}$ is a linear differential operator with constant coefficients, and Eq. (2.11) then has a local form. More generally, the hydrodynamic modes for nonequilibrium states will involve a coupling of different wave vectors. This new feature of the linear Langevin equations for nonequilibrium fluctuations will be referred to as linear mode coupling. It is to be distinguished from the mode coupling due to nonlinearities in z_α which have been neglected in Eq. (2.11).

III. HYDRODYNAMIC MODES FOR SHEAR FLOW

The macroscopic state of uniform shear flow is characterized by an average velocity field

$$v_{0i}(\mathbf{r},t) = a_{ij} r_j \ ,$$
$$a_{ij} \equiv a\delta_{ix}\delta_{jy} \tag{3.1}$$

corresponding to a flow along the x axis with a constant gradient a along the y axis. An arbitrary additive constant velocity is sometimes included but can be removed by a suitable Galilean transformation. This velocity field is a solution to the macroscopic Navier-Stokes equations for spatially uniform pressure and macroscopic internal energy density ε_0 determined from the energy equation

$$\frac{\partial \varepsilon_0}{\partial t} = \lambda \frac{\partial^2 T_0}{\partial y^2} + \eta a^2 \ . \tag{3.2}$$

Here λ and η are the thermal conductivity and shear viscosity, and T_0 is the macroscopic temperature. There are two different solutions to Eq. (3.2) of interest. The first is homogeneous shear flow, for which the temperature and density are spatially uniform but the energy increases according to

$$\frac{\partial \varepsilon_0(t)}{\partial t} = \eta a^2 \ . \tag{3.3}$$

The conditions of homogeneous shear are most closely related to the Lees-Edwards conditions for computer simulations.[13] A second solution exists for time-independent properties, but with spatially varying temperature,

$$T_0(y) = T_1 - \frac{\eta a^2}{2\lambda} y^2 \ , \tag{3.4}$$

where T_1 is a constant determined from the boundary condition that the temperature at the surfaces is constant. In the following the conditions of homogeneous shear flow will be assumed for definiteness. However, as observed below, the results obtained will be valid for both cases (3.3) and (3.4).

It is now straightforward to obtain the linear equations (2.11) for the case chosen. The form of these equations is simplified considerably by a suitable choice of variables,

$$\tilde{z}(\mathbf{k},t) \leftrightarrow (c_1 \delta\tilde{\rho}(\mathbf{k},t),\, c_2 \delta\tilde{\varepsilon}(\mathbf{k},t),\, \hat{\mathbf{e}}^{(\alpha)}(\mathbf{k})\cdot\delta\tilde{\mathbf{v}}(\mathbf{k},t)) \ , \tag{3.5}$$

$$c_1^2 = \rho_0^{-2} \left[\frac{\partial p_0}{\partial \rho_0} \right]_{\varepsilon_0} , \quad c_2^2 = (\rho_0 h_0)^{-1} \left[\frac{\partial p_0}{\partial \varepsilon_0} \right]_{\rho_0} , \tag{3.6}$$

where $h_0 = \varepsilon_0 + p_0$ is the average enthalpy density, and

$$\delta\rho=\rho-\rho_0,\quad \delta\varepsilon=u'-\varepsilon_0,\quad \delta\mathbf{v}=\mathbf{v}-\mathbf{v}_0\ . \tag{3.7}$$

The vectors $\{\hat{\mathbf{e}}^{(\alpha)}(k)\}$ are a set of three pairwise orthogonal unit vectors with $\hat{\mathbf{e}}^{(1)}(k)$ along $\mathbf{k}$. The linear Langevin equations for these variables are found to be

$$\left[\frac{\partial}{\partial t}-a_{ij}k_i\frac{\partial}{\partial k_j}\right]\tilde{z}_\alpha(\mathbf{k},t)+L_{\alpha\beta}(\mathbf{k},a,t)\tilde{z}_\beta(\mathbf{k},t)=\overline{R}_\alpha(\mathbf{k},t)\ . \tag{3.8}$$

The detailed forms of $\overline{R}_\alpha(k,t)$ and $L_{\alpha\beta}(\mathbf{k},a,t)$ are given in Appendix A. The linear mode coupling is represented by the differential operator with respect to $\mathbf{k}$, a simplification of Eq. (2.11) due to the linearity of the imposed velocity field. The matrix $L_{\alpha\beta}(\mathbf{k},a,t)$ also depends on the shear rate, but in a manner that does not couple modes of different wave vector, $\mathbf{k}$. Finally, the time dependence of $L_{\alpha\beta}(\mathbf{k},a,t)$ is due entirely to the viscous heating expressed by Eq. (3.3).

Since the irreversible fluxes at Navier-Stokes order are valid only to first order in the gradients, this matrix is valid only to order k^2. In the present context it is necessary to specify the relative magnitude of the shear rate a as well. To motivate the choice, it is noted that there will be secular terms in the solution to Eq. (3.8) of the form (at). Also, heat and momentum diffusion and sound damping occur on a time scale of order $\nu_0k^2t\sim 1$, where $\nu_0\equiv\eta_B/\rho_0$ is the kinematic viscosity. Therefore, to investigate shear-rate effects on the same time scale as hydrodynamic dissipation, the shear rate is chosen to satisfy

$$a\lesssim\nu_0k^2\ . \tag{3.9}$$

The wave vector must be small compared to the mean free path l for the validity of the Navier-Stokes approximation so that the condition (3.9) becomes $a\ll\nu_0/l^2$. This is a relatively weak constraint and allows for quite large shear rates (e.g., for air at STP the restriction is $a\ll 10^9\ \mathrm{sec}^{-1}$). With this choice the time dependence of $L_{\alpha\beta}(\mathbf{k},a,t)$ may be neglected, since it generates nonsecular contributions of order $ak\lesssim\nu_0k^3$.

A further simplification of Eq. (3.8) is possible by a transformation to diagonal form in terms of a set of five linearly independent vectors $\zeta^{(i)}$ that are solutions to the generalized eigenvalue problem

$$\left[-a_{ij}k_i\frac{\partial}{\partial k_j}+L\right]\zeta^{(i)}=\lambda_i\zeta^{(i)}\ . \tag{3.10}$$

The eigenvectors $\zeta^{(i)}$ and eigenvalues λ_i can be obtained explicitly in the context of perturbation theory, using k as a small parameter. Since the matrix L is not Hermitian it is also necessary to introduce an associated biorthogonal set of vectors $\eta^{(i)}$ with the property

$$(\eta^{(i)},\zeta^{(j)})\equiv\sum_{\alpha=1}^{5}\eta_\alpha^{(i)*}\zeta_\beta^{(j)}=\delta_{ij}\ . \tag{3.11}$$

The first equality of (3.11) defines the scalar product in the five-dimensional space of hydrodynamic variables. The determination of the eigenvectors and eigenvalues is described in Appendix B. In particular, the eigenvalues to order k^2 are

$$\begin{aligned}
\lambda_1(k,a)&=-ic_0k+\tfrac{1}{2}(\Gamma_0k^2+ak_xk_y/k^2)\ ,\\
\lambda_2(k,a)&=+ic_0k+\tfrac{1}{2}(\Gamma_0k^2+ak_xk_y/k^2)\ ,\\
\lambda_3(k,a)&=D_{0T}k^2\ ,\\
\lambda_4(k,a)&=\nu_0k^2-ak_xk_y/k^2\ ,\\
\lambda_5(k,a)&=\nu_0k^2\ .
\end{aligned} \tag{3.12}$$

Here c_0 is the sound speed, Γ_0 is the sound damping constant, and D_{0T} is the thermal diffusivity. For zero shear rate the first two eigenvalues represent damped sound propagation and the third eigenvalue is for thermal diffusion, while the last two describe transverse momentum (shear) diffusion. The effect of the nonequilibrium state in (3.12) is then simply to shift the sound damping by a term proportional to the shear rate and to remove the degeneracy of the shear modes.

The general solution to the Langevin equations (3.8) can now be constructed from these eigenvectors and eigenvalues in the form

$$\begin{aligned}
\tilde{z}_\alpha(k,t)=&\int\frac{d\mathbf{k}'}{(2\pi)^3}G_{\alpha\beta}(\mathbf{k},\mathbf{k}';t-t_0)\tilde{z}_\beta(\mathbf{k}',t_0)\\
&+\int_{t_0}^{t}d\tau\int\frac{d\mathbf{k}'}{(2\pi)^3}G_{\alpha\beta}(\mathbf{k},\mathbf{k}';t-\tau)\overline{R}_\beta(\mathbf{k}',\tau)\ .
\end{aligned} \tag{3.13}$$

The Green's function $G_{\alpha\beta}(\mathbf{k},\mathbf{k}';t)$ has the representation

$$G_{\alpha\beta}(\mathbf{k},\mathbf{k}';t)=\sum_{i=1}^{5}\zeta_\alpha^{(i)}(\mathbf{k})\eta_\beta^{(i)}(\mathbf{k}')G^{(i)}(\mathbf{k},\mathbf{k}';t)\ , \tag{3.14}$$

where $G^{(i)}(\mathbf{k},\mathbf{k}';t)$ satisfies

$$\begin{aligned}
&\left[\frac{\partial}{\partial t}-a_{ij}k_i\frac{\partial}{\partial k_j}+\lambda_i(\mathbf{k},a)\right]G^{(i)}(\mathbf{k},\mathbf{k}';t)=0\ ,\\
&G^{(i)}(\mathbf{k},\mathbf{k}';0)=(2\pi)^3\delta(\mathbf{k}-\mathbf{k}')\ .
\end{aligned} \tag{3.15}$$

The form (3.14) allows identification of the five linearly independent excitations possible in the fluid under shear. Accordingly, the hydrodynamic modes are the solutions to Eqs. (3.15). It is important to note the role of linear mode coupling in these equations. If the latter is neglected, the resulting approximate modes are entirely characterized by the eigenvalues $\lambda_i(\mathbf{k},a)$. Since the effect of the shear rate in Eqs. (3.12) is to change the sound damping by a term of equal magnitude but opposite sign to that of the shear mode, one of the modes will necessarily be unstable at sufficiently large shear rate. However, this conclusion is not justified since inclusion of the linear mode coupling in (3.15) stabilizes the modes at all shear rates, as demonstrated below. The solutions are readily found to be

$$\begin{aligned}
&G^{(i)}(\mathbf{k},\mathbf{k}';t)=(2\pi)^3\delta(\mathbf{k}-\mathbf{k}'(t))E^{(i)}(\mathbf{k},t)\ ,\\
&E^{(i)}(\mathbf{k},t)=\exp\left[-\int_0^t d\tau\,\lambda_i(k(-\tau))\right]\ .
\end{aligned} \tag{3.16}$$

Here $k_i(t)=k_i-k_j a_{ji} t$.

These results allow the Green's function, (3.14), to be put in the form, for $t \geq t'$,

$$G_{\alpha\beta}(\mathbf{k},\mathbf{k}';t)=(2\pi)^3\delta(\mathbf{k}-\mathbf{k}'(t))\sum_{i=1}^{5}\zeta_\alpha^{(i)}(\mathbf{k},t)\eta_\beta^{(i)}(\mathbf{k}') , \quad (3.17)$$

where the time-dependent vector $\zeta^{(i)}(\mathbf{k},t)$ is

$$\zeta^{(i)}(\mathbf{k},t)=E^{(i)}(\mathbf{k},t)\xi^{(i)}(\mathbf{k}) . \quad (3.18)$$

These five linearly independent vectors are defined to be the hydrodynamic modes for small deviations from uniform shear flow. They are solutions to the equations

$$\left[\frac{\partial}{\partial t}+\lambda_i(k(-t),a)\right]\zeta^{(i)}(\mathbf{k},t)=0 . \quad (3.19)$$

These equations have a simple interpretation. They result from Fourier transformation of the Navier-Stokes equations (2.10) referred to the Lagrangian coordinate frame. The latter is obtained from the transformation

$$r_i'=r_i-v_{0i}(r)t=r_i-a_{ij}r_j t . \quad (3.20)$$

Effectively, this induces a local Galilean transformation that brings each fluid element to rest relative to the macroscopic motion, and it is quite natural that the hydrodynamic modes have the simple form of equilibriumlike excitations in this frame. However, the wave-vector dependence of the eigenvalues λ_i in this frame is necessarily time dependent. This latter effect is responsible for a qualitative difference of the $E^{(i)}(k,t)$ from simple exponential functions of t. (This time-dependent wave vector may be interpreted as the Fourier transform variable for spatial functions referred to the local Lagrangian frame for the fluid under shear flow, i.e., $r_i'=r_i-a_{ij}r_j t$.) The specific forms of the functions $E^{(i)}(\mathbf{k},t)$ are

$$\begin{aligned}
E^{(1)}(\mathbf{k},t)&=[k/k(-t)]^{1/2}\exp[-ic_0 k\alpha(t) \\
&\qquad -\tfrac{1}{2}\Gamma_0 k^2\beta(t)] , \\
E^{(2)}(\mathbf{k},t)&=[E^{(1)}(\mathbf{k},t)]^* , \\
E^{(3)}(\mathbf{k},t)&=\exp[-D_{0T}k^2\beta(t)] , \\
E^{(4)}(\mathbf{k},t)&=[k(-t)/k]\exp[-\nu_0 k^2\beta(t)] , \\
E^{(5)}(\mathbf{k},t)&=[k/k(-t)]E^{(4)}(\mathbf{k},t) .
\end{aligned} \quad (3.21)$$

The time-dependent functions $\alpha(t)$ and $\beta(t)$ are

$$\alpha(t)\equiv(2ak_x k)^{-1}\times\left[[k_y(-t)k(-t)-k_y k]+k_\perp^2\ln\left[\frac{k(-t)+k_y(-t)\mathrm{sgn}(ak_x)}{k+k_y\mathrm{sgn}(ak_x)}\right]\right] , \quad (3.22)$$

$$\beta(t)\equiv(ak_x k^2)^{-1}\{k_\perp^2[k_y(-t)-k_y]+\tfrac{1}{3}[k_y^3(-t)-k_y^3]\} , \quad k_\perp^2\equiv k^2-k_y^2 .$$

In particular, it is seen that for large t,

$$\alpha(t)\rightarrow(ak_x/2k)t^2, \quad \beta(t)\rightarrow\tfrac{1}{3}(ak_x/k)^2 t^3 . \quad (3.23)$$

Since the magnitude of $\beta(t)$ is positive the functions $E^{(i)}(\mathbf{k},t)$ decay (for $t>0$) and hence represent stable excitations.

To illustrate the effects of strong shear flow on the hydrodynamic modes, consider the special case of an initial sound excitation along the direction of flow, i.e., $\tilde{z}_\alpha(\mathbf{k},0)=\xi_\alpha^{(1)}(k)\delta(\mathbf{k}-\mathbf{k}_0)$ with $\mathbf{k}_0\equiv k_0\hat{\mathbf{x}}$. Then the average of (3.13) gives[37]

$$\begin{aligned}
\langle\langle\tilde{z}_\alpha(\mathbf{k},t)\rangle\rangle&=G_{\alpha\beta}(\mathbf{k},\mathbf{k}_0;t)\xi_\beta^{(1)}(\mathbf{k}_0)/(2\pi)^3 \\
&=\delta(\mathbf{k}-\mathbf{k}_0(t))\xi_\alpha^{(1)}(k)E^{(1)}(\mathbf{k},t) ,
\end{aligned} \quad (3.24)$$

where the second equality follows from Eqs. (3.17) and (3.18). Inverting the Fourier transform in Eq. (3.24) gives the space-time evolution of this initial excitation,

$$\langle\langle z_\alpha(\mathbf{r},t)\rangle\rangle=\xi_\alpha^{(1)}(\mathbf{k}_0(t))(1+a^2t^2)^{1/4}e^{-\Gamma_0 k_0^2\beta(t;-a)/2}\times\cos\{k_0[x(t)-c_0\alpha(t;-a)]\} , \quad (3.25)$$

where $\alpha(t;a)$ and $\beta(t;a)$ are defined by Eq. (3.23), and $x(t)$ is the Lagrangian coordinate of Eq. (3.20), $x(t)\equiv x+ayt$. The corresponding result for an excitation in an equilibrium fluid results from the replacements, $at=0$ and $\alpha(t)=\beta(t)=1$. The expected effects of flow resulting from a Doppler shift of the velocity are contained in $x(t)$. These may be suppressed by restricting attention to the time variations in the plane $y=0$. Since all points in this plane are stagnation points, the nonequilibrium effects on sound propagation and damping are due to *gradients* of the velocity field. Without loss of generality, $x(0)$ is also chosen to be zero. If the time is measured in units of $(\Gamma_0 k_0^2)^{-1}$, the relevant parameters of (3.25) are $(c_0k_0/\Gamma_0k_0^2)$ and $(a/\Gamma_0k_0^2)$. Choosing a typical value of $(c_0k_0/\Gamma_0k_0^2)=10^{-1}$, Fig. 1 shows the results for $(a/\Gamma_0k_0^2)=0$ and 1. The effects of shear flow are twofold. First, the effective sound velocity is approximately

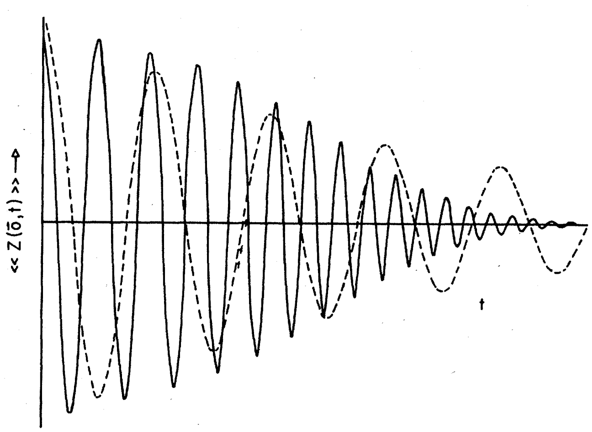

FIG. 1. Propagation and damping of sound wave in shear flow for $a/\Gamma_0k_0^2=1$ (——) and for $a/\Gamma_0k_0^2=0$ (- - -).

twice that for zero shear rate on this time scale. In addition, the attenuation is significantly enhanced by the shear rate after about five cycles. At much larger times the period varies to a greater extent, but the amplitude is then very small. Although $(a/\Gamma_0 k_0^2)=1$ would be difficult to attain for simple atomic fluids, it is likely that such modifications of sound and other hydrodynamic modes could be observed in driven colloidal suspensions under shear flow.[22,23]

IV. CORRELATION FUNCTIONS

The correlation functions for the deviation of the hydrodynamic variables from the macroscopic state of uniform shear flow are defined by

$$C_{\alpha\beta}(\mathbf{k},t;\mathbf{k}',t';a)\equiv\langle\tilde{z}_\alpha(\mathbf{k},t)\tilde{z}_\beta(\mathbf{k}',t')\rangle\ , \qquad (4.1)$$

where the angle brackets denote the nonequilibrium average, and the shear rate dependence has been made explicit on the left side of (4.1). Time-reversal invariance and stationarity impose the symmetries

$$C_{\alpha\beta}(\mathbf{k},t;\mathbf{k}',t';a)=C_{\alpha\beta}(\mathbf{k},t-t';\mathbf{k}',0;a)\ ,$$
$$C_{\alpha\beta}(\mathbf{k},t;\mathbf{k}',t';a)=P_\alpha P_\beta C^*_{\beta\alpha}(\mathbf{k}',t;\mathbf{k},t';-a)\ , \qquad (4.2)$$

where $P_\alpha=\pm1$ denotes the parity of $\tilde{z}_\alpha$ under time reversal. It is therefore sufficient to consider $C_{\alpha\beta}(\mathbf{k},t;\mathbf{k}';0;a)$ for $t\geqq0$, without loss of generality. To calculate the correlation function, Eq. (3.13) for $t_0\to-\infty$ may be used,

$$z_\alpha(\mathbf{k},t)=\int_{-\infty}^{t}d\tau\sum_{i=1}^{5}\zeta_\alpha^{(i)}(\mathbf{k},t-\tau)F^{(i)}(\mathbf{k}(\tau-t);\tau)\ , \qquad (4.3)$$

where the effective random force $F^{(i)}$ is

$$F^{(i)}(\mathbf{k},t)\equiv\eta_\beta^{(i)}(\mathbf{k})\overline{R}_\beta(\mathbf{k},t)\ . \qquad (4.4)$$

The correlation function is then, for $t\geqq0$,

$$C_{\alpha\beta}(\mathbf{k},t;\mathbf{k}',0;a)=\sum_{i,j}\int_0^\infty d\tau\int_0^\infty d\tau'\zeta_\alpha^{(i)}(\mathbf{k},\tau)\zeta_\beta^{(j)}(\mathbf{k}',\tau')\langle F^{(i)}(\mathbf{k}(-\tau),t-\tau)F^{(j)}(\mathbf{k}'(-\tau'),-\tau')\rangle\ . \qquad (4.5)$$

The properties (A3) for the random variables imply

$$\langle F^{(i)}(\mathbf{k},t)F^{j}(\mathbf{k}',t')\rangle=(2\pi)^3\delta(\mathbf{k}+\mathbf{k}')\delta(t-t')F^{(ij)}(\mathbf{k})\ , \qquad (4.6)$$

which defines the matrix $F^{(ij)}(\mathbf{k})$. Use of (4.6) in (4.5) then gives an expansion for the correlation function in terms of the hydrodynamic modes,

$$C_{\alpha\beta}(\mathbf{k},t;\mathbf{k}',0;a)=(2\pi)^3\delta(\mathbf{k}+\mathbf{k}'(t))\times\sum_{i=1}^{5}\zeta_\alpha^{(i)}(\mathbf{k},t)\eta_\sigma^{(i)}(-\mathbf{k}')C_{\sigma\beta}(-\mathbf{k}';a)\ . \qquad (4.7)$$

The coefficients in this expansion $C_{\alpha\beta}(\mathbf{k}';a)$ are related to the equal-time correlation functions by

$$C_{\alpha\beta}(\mathbf{k},0;\mathbf{k}',0;a)=(2\pi)^3\delta(\mathbf{k}+\mathbf{k}')C_{\alpha\beta}(\mathbf{k};a)\ . \qquad (4.8)$$

The explicit form of these coefficients, as determined from (4.5) and (4.6), is

$$C_{\alpha\beta}(\mathbf{k};a)=\int_0^\infty d\tau\sum_{i,j}\zeta_\alpha^{(i)}(\mathbf{k},\tau)\zeta_\beta^{(j)}(-\mathbf{k},\tau)F^{(ij)}(\mathbf{k}(-\tau))\ . \qquad (4.9)$$

Equations (4.7)–(4.9) are the primary results of this section. The first shows that the time-dependent correlations decay according to the same linear laws as those for small deviations of the hydrodynamic variables from the macroscopic state of shear flow. This represents a generalization of Onsager's assumption on the regression of fluctuations to nonequilibrium states.[5,6,35] Equation (4.9) shows that the equal-time correlation functions are determined from the noise amplitude and the hydrodynamic modes. These results have a more familiar form in terms of the differential equations they satisfy, as follows directly from (4.7)–(4.9),

$$\left[\frac{\partial}{\partial t}-a_{ij}k_i\frac{\partial}{\partial k_j}\right]C_{\alpha\beta}(\mathbf{k},t;\mathbf{k}',t')+L_{\alpha\sigma}(k,a)C_{\sigma\beta}(\mathbf{k},t;\mathbf{k}',t')=0\ , \qquad (4.10)$$

$$\left[-\delta_{\alpha\sigma}a_{ij}k_i\frac{\partial}{\partial k_j}+L_{\alpha\sigma}(k,a)\right]C_{\sigma\beta}(\mathbf{k};a)+[L_{\beta\sigma}(-k,a)]C_{\alpha\sigma}(\mathbf{k};a)=R_{\alpha\beta}(\mathbf{k})\ , \qquad (4.11)$$

where $R_{\alpha\beta}(\mathbf{k})$ is the amplitude of the random forces,

$$\langle\overline{R}_\alpha(\mathbf{k},t)\overline{R}_\beta(\mathbf{k}',t')\rangle\equiv\delta(\mathbf{k}+\mathbf{k}')\delta(t-t')R_{\alpha\beta}(\mathbf{k})\ . \qquad (4.12)$$

Equation (4.10) is the linear regression law, while Eq. (4.11) may be identified as a nonequilibrium fluctuation-dissipation relation. These equations show the close relationship of the dynamics of fluctuations to the macroscopic dynamics, even for states far from equilibrium.

The equal-time correlation functions $C_{\alpha\beta}(\mathbf{k};a)$ are given in Appendix C [Eqs. (C8) and (C9)]. One interesting feature of these functions is that some of them are long ranged. For example, the inverse transform of the density-density correlation function defined by

$$C_{\rho\rho}(\mathbf{r})=\int\frac{d\mathbf{k}}{(2\pi)^3}e^{i\mathbf{k}\cdot\mathbf{r}}C_{\rho\rho}(\mathbf{k};a)$$

is easily shown to have the form

$$C_{\rho\rho}(\mathbf{r})=l^{-3}k_BT_0\rho_0^3\chi_T[\delta(\mathbf{r}/l)+(l/r)F(\mathbf{r}/l)]\ , \qquad (4.13)$$

where $l=(\Gamma_0/a)^{1/2}$ is a characteristic length associated with the shear rate. The first term in the square brackets of Eq. (4.13) is the usual result for equilibrium fluctuations and expresses the idealized conditions of δ-function correlations introduced in the Langevin forces (A3). More accurately, of course, there exist equilibrium density correlations of finite range on the scale of the interatomic force range. The second term in (4.13) is quite different and represents long-range correlations originating entirely from the nonuniformity of the fluid under shear flow. The factor of (l/r) in this term has been obtained previously by direct expansion of $C_{\rho\rho}(\mathbf{r})$ to first order in the shear rate. Equation (4.13) shows that such an expansion is valid for $r/l \ll 1$, with a leading coefficient of $F(0) = -a_{ij}r_ir_j/8\pi\gamma a r^2$ where $\gamma = C_p/C_v$ is the ratio of specific heats. Although this (l/r) dependence is certainly long ranged compared to the equilibrium correlation function, it is seen to be the short-range limit of the second term in (4.13). To study the behavior of this function for larger (r/l) is a straightforward but multidimensional numerical problem. Instead, the related, but somewhat simpler, problem of the density correlations between a plane orthogonal to the flow and a point on the x axis is considered,

$$\int dy \int dz\, C_{\rho\rho}(\mathbf{r}) \equiv l^{-1}k_BT_0\rho_0^3\chi_T[\delta(x/l)+\gamma H(x/l)] \; . \quad (4.14)$$

The nonequilibrium contribution $H(x/l)$ is reduced in this case to the simple one-dimensional integral

$$H(x/l) = (2\sqrt{\pi})^{-1}\int_0^\infty dt\,(1+t^2)^{-3/2}t^{-1/2}(1+\tfrac{1}{3}t^2)^{-1/2} \times \exp\{-(x/l)^2[4t(1+\tfrac{1}{3}t^2)]^{-1}\} \; . \quad (4.15)$$

For large (x/l) the asymptotic decay of correlations along the flow is algebraic:

$$H(x/l) \to 0.828(x/l)^{-5/3} \; . \quad (4.16)$$

At very short distances these one-dimensional correlations are approximately Gaussian. The transition between these limits is shown in Fig. 2. If the conditions of Fig. 1 for the sound excitation are assumed then the wavelength of the sound wave, $\lambda \sim 2\pi/k_0$, corresponds to $(x/l) \sim 2\pi$. Therefore, under these conditions of strong shear flow the time-dependent density fluctuations will not only exhibit the temporal modifications illustrated in Fig. 1, but also the slow spatial decay of (4.16). The former is simply an effect of macroscopic hydrodynamics, where the latter is attributed to nonequilibrium fluctuations.

The time correlation functions may be used to compute the dynamic structure factor which describes the intensity of light scattered from the system,

$$S(\mathbf{k},\omega) \equiv \int_{-\infty}^{\infty} dt \int\int \frac{d\mathbf{k}_1 d\mathbf{k}_2}{(2\pi)^6}\tilde{\theta}(\mathbf{k}-\mathbf{k}_1)\tilde{\theta}(\mathbf{k}+\mathbf{k}_2) \times C_{\rho\rho}(\mathbf{k}_1,t;\mathbf{k}_2,0;a)e^{i\omega t} \; , \quad (4.17)$$

where $\tilde{\theta}(\mathbf{k})$ is the Fourier transform of a normalized form factor designed to restrict spatial integrals to the volume irradiated. This expression is simplified in Appendix C to identify the modified Brillouin and Rayleigh peaks,

$$S(\mathbf{k};\omega) = S_R(\mathbf{k};\omega) + S_B(\mathbf{k};\omega) + S_B(\mathbf{k};-\omega) \; , \quad (4.18)$$

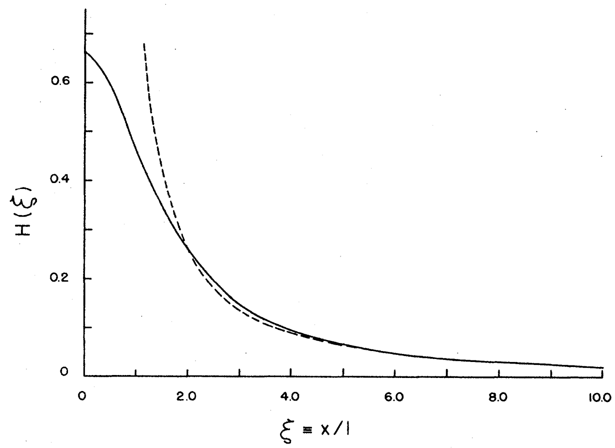

FIG. 2. Spatial correlations of density along direction of flow for $a/\Gamma_0k_0^2=1$ (——) and corresponding asymptotic form, Eq. (4.16) (- - -).

with

$$S_B(\mathbf{k};\omega) = 2R_e\left[-i(\omega+ck)+\tfrac{1}{2}(\Gamma_0k^2+ak_xk_y/k^2) - \tfrac{1}{2}a_{ij}k_i\frac{\partial}{\partial k_j}\right]^{-1} \times c_1^{-2}\xi_1^{(1)}\eta_\alpha^{(1)}C_{\alpha 1}(k;a) \; ,$$

$$S_R(\mathbf{k};\omega) = 2R_e\left[-i\omega + D_{0T}k^2 - \tfrac{1}{2}a_{ij}k_i\frac{\partial}{\partial k_j}\right]^{-1} \times c_i^{-2}\xi_1^{(3)}\eta_\alpha^{(3)}C_{\alpha 1}(k;a) \; . \quad (4.19)$$

These are not simply Lorentzians, due to the operator character of the terms in the large parentheses of Eqs. (4.19). Expansion of (4.19) to first order in shear rate gives agreement with earlier results.[5-8] The shape of these lines at large shear rates is straightforward, but numerically difficult, to obtain from (4.19) except for selected directions of the scattered wave vector $\mathbf{k}$.[12] However, the Landau-Placzek coefficient, defined as the ratio of the integrated intensities of the Rayleigh and Brillouin peaks, can be obtained directly:

$$\mathscr{R} \equiv \int d\omega\, S_R(\mathbf{k};\omega) \Big/ \int d\omega[S_B(\mathbf{k};\omega)+S_B(\mathbf{k};-\omega)]$$
$$= \xi_1^{(3)}\eta_\alpha^{(3)}C_{\alpha 1}(k;a)/\xi_1^{(1)}\eta_\alpha^{(1)}C_{\alpha 1}(k;a)$$
$$= [C_{\rho\rho}-(\rho_0/h_0)C_{\varepsilon\rho}]/[\alpha^2C_{\rho\rho}+(\rho_0/h_0)C_{\varepsilon\rho}] \; , \quad (4.20)$$

where $\alpha = \rho_0c_1/h_0c_2$. The Rayleigh intensity in the numerator is easily obtained from the first two equations of (C8) in Appendix C,

$$\int d\omega\, S_R(\mathbf{k};\omega) = (\gamma-1)/\gamma \; , \quad (4.21)$$

where γ again denotes the ratio of specific heats. This is simply the equilibrium result, which is now seen to hold to *all orders* in the shear rate. Similarly, the intensity of the Brillouin zone peaks can be obtained from Eqs. (C8), and the resulting Landau-Placzek ratio is

$$\mathscr{R}=(\gamma-1)/[1+\gamma\Delta_1(\mathbf{k};a)] \ . \tag{4.22}$$

Here $\Delta_1(\mathbf{k};a)$ is the nonequilibrium part of the equal-time density autocorrelation function. Generally, this ratio is a function of the magnitude and direction of the wave vector $\mathbf{k}$ as well as the shear rate. These effects are illustrated for large shear rate ($a/\Gamma_0 k^2=1$) in Fig. 3, where $\mathbf{k}$ is taken in the x-y plane and the deviation $R=[\mathscr{R}(a)-\mathscr{R}(0)]/\mathscr{R}(0)$ is evaluated as a function of the angle between $\mathbf{k}$ and the x axis.

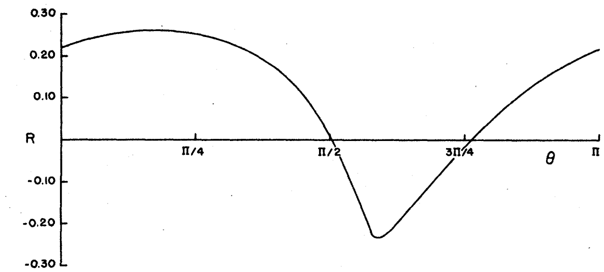

FIG. 3. Deviation of the Landau-Placzek ratio, Eq. (4.22), as a function of angles for $a/\Gamma_0 k^2=1$ and $k_z=0$. The value at $\theta=\pi/2$ corresponds to $k_x=0$, for which the equilibrium result holds.

V. PROBABILITY DENSITIES

The correlation functions (4.1) were evaluated simply and directly from the Langevin equations. However, to calculate higher-order correlation functions or averages of more general functions of the $\{\tilde{z}_\alpha\}$ it is useful to construct the probability densities for these variables. For the Gaussian-Markovian process considered here it is sufficient to know the probability and joint probability densities, defined by

$$\begin{aligned} P(z;t) &\equiv \langle \delta(\tilde{z}(t)-z)\rangle \ , \\ P(z,t;z',t') &\equiv \langle \delta(\tilde{z}(t)-z)\delta(\tilde{z}(t')-z')\rangle \ . \end{aligned} \tag{5.1}$$

The angle brackets denote an average over the random variables $\{\overline{R}_\alpha\}$ and over a specified ensemble of initial values for $\{\tilde{z}_\alpha(0)\}$. The δ function in (5.2) is an abbreviated notation for

$$\delta(\tilde{z}(t)-z)\equiv \prod_{\alpha,\mathbf{k}} \delta(\tilde{z}_\alpha(\mathbf{k},t)-z_\alpha(\mathbf{k})) \ . \tag{5.2}$$

These probability densities are most easily constructed from the conditional probability density

$$W(z,t;z',t')\equiv P(z,t;z't')/P(z',t') \ . \tag{5.3}$$

Since the process is Markovian, the joint probability density is independent of the ensemble of initial values, $\{\tilde{z}_\alpha(0)\}$. Furthermore, with condition (3.11) the process is approximately stationary and $W(z,t;z',t')=W(z,t-t';z',0)$. Therefore, it is sufficient to calculate

$$W(z,t';z',0)=\langle \delta(\tilde{z}(t\,|\,z')-z)\rangle_{\mathbf{R}} \ , \tag{5.4}$$

where the angle brackets now refer to an average over only the random components R, and $\tilde{z}(t\,|\,z')$ is the solution to the Langevin equations with initial value $\tilde{z}(0\,|\,z')=z'$. The latter is given by Eq. (3.13), written in the abbreviated notation

$$\tilde{z}(t\,|\,z')=G(t)z'+\int_0^t d\tau\, G(t-\tau)\overline{R}(\tau) \ . \tag{5.5}$$

An integral representation for the δ function in (5.4) and use of (5.5) then gives

$$\begin{aligned} W(z,t;z',0) &= \int d\lambda\, e^{i\lambda\cdot[z-G(t)z']}\left\langle \exp\left[-i\lambda\cdot\int_0^t d\tau\, G(t-\tau)\overline{R}(\tau)\right]\right\rangle_{\overline{R}} \\ &= \int d\lambda \exp\{i\lambda[z-G(t)z']-\lambda^2 M(t)\} \\ &= \{\det[\pi M(t)]\}^{-1/2}\exp\{-\tfrac{1}{2}[z-G(t)z']\cdot M^{-1}(t)\cdot[z-G(t)z']\} \ , \end{aligned}$$

where the second equality is obtained from the first from the Gaussian statistics for $\overline{R}(t)$, and the matrix $M(t)$ is defined by

$$M(t)=\int_0^t d\tau \int_0^t d\tau' G(t-\tau)\cdot\langle \mathbf{R}(\tau)\mathbf{R}(\tau')\rangle\cdot G^T(t-\tau') \ . \tag{5.7}$$

Use of the Onsager regression equation and fluctuation-dissipation relation, Eqs. (4.10) and (4.11), allows (5.7) to be written more explicitly in terms of the steady-state correlation functions of Sec. IV,

$$M(t)=C(0)-C(t)\cdot C^{-1}(0)\cdot C^T(t) \ . \tag{5.8}$$

Here $C(t)$ is the abbreviated notation for the correlation function $C_{\alpha\beta}(k,t;k',0)$.

The probability and joint probability densities are now obtained simply from the conditional probability density,

$$\begin{aligned} P(z,t) &= \int dz' W(z,t;z',0)P(z',0) \ , \\ P(z,t;z',t') &= W(z,t;z',t')P(z',t') \ , \end{aligned} \tag{5.9}$$

where $P(z',0)$ is the specified initial density. Since the conditional probability density is specified entirely in terms of the correlation functions calculated in Sec. IV, they provide through (5.9) complete information for any desired average. Some important conclusions follow from the explicit form of the conditional probability density. For example, the stationary state exists as a consequence of the stability of the correlation functions and is given by

$$\begin{aligned} P_{ss}(z) &\equiv \lim_{t\to\infty} P(z,t) \\ &= \{\det[\pi C(0)]\}^{-1/2}\exp[-\tfrac{1}{2}z\cdot C^{-1}(0)\cdot z] \ . \end{aligned} \tag{5.10}$$

As expected for a linear Langevin equation with Gaussian random components, the variables $\{z_\alpha\}$ also have a Gaussian distribution in the stationary state, with covariance determined from the nonequilibrium equal-time fluctuations. Equation (5.10) is actually a stronger result, implying that $P_{ss}(z)$ is not only stationary but also represents the asymptotic state for a wide class of initial conditions. It also follows from Eqs. (5.6), (5.9), and (5.10) that the joint probability density is Gaussian in the steady state.

The matrix $C^{-1}(0)$ is easily inverted using the results (C8) of Appendix C, but will not be given here. Instead, to illustrate (5.10) in more detail, the reduced stationary distribution for the longitudinal components of the velocity field, $z_3=\hat{\mathbf{k}}\cdot\delta\mathbf{v}(k)\equiv u(k)$, can be written down from inspection since $C_{33}(\mathbf{k};a)$ does not couple to the other correlation functions. The result can be expressed in the suggestive form

$$P_{ss}(u(k))=[2\pi m k_B T(\mathbf{k},a)]^{-1/2}\times\exp\{-m\,|u(k)|^2/[2k_B T(\mathbf{k},a)]\}\ , \qquad (5.11)$$

where the effective "temperature" is

$$T(\mathbf{k},a)\equiv T_0 R(0;0)/R(\mathbf{k};a) \qquad (5.12)$$

and $R(\mathbf{k};a)$ is the Landau-Placzek ratio calculated in Sec. IV.

The above results could have been obtained also from the linear Fokker-Planck equation associated with the Langevin equations (3.7). The form of the Fokker-Planck equation is easily obtained from direct differentiation of (5.9) with respect to time,

$$\frac{\partial}{\partial t}P(z,t)=-\sum_k \frac{\partial}{\partial z_\alpha(k)}[J_{0\alpha}(z,t)+J_{1\alpha}(z,t)]\ , \qquad (5.13)$$

where $J_{0\alpha}(z,t)$ is the deterministic or "drift" contribution to the probability current density,

$$J_{0\alpha}(z,t)\equiv\left[a_{ij}k_i\frac{\partial}{\partial k_j}z_\alpha(k)-L_{\alpha\beta}(k,a)z_\beta(k)\right]P(z,t)\ , \qquad (5.14)$$

and $J_{1\alpha}(z,t)$ is the "diffusion" component of the current density,

$$J_{1\alpha}(z,t)=\tfrac{1}{2}R_{\alpha\beta}(k)\frac{\partial}{\partial z_\beta(-k)}P(z,t)\ . \qquad (5.15)$$

The diffusion tensor $R_{\alpha\beta}(k)$ is the amplitude of the correlation matrix for the random components in the Langevin equations, as defined in (4.11). Equations (5.13)–(5.15) are the expected results for the relationship of Fokker-Planck and Langevin descriptions.[38]

VI. DISCUSSION

The effect of the macroscopic state of uniform shear flow is to modify the evolution of small fluctuations around the average nonequilibrium steady-state values of the hydrodynamic parameters. This hydrodynamic effect occurs, at the level of the linearized evolution equations, because of the coupling between the fluctuations and the macroscopic flow field generated by the Oseen terms. Two modifications result. The first of these is the appearance of a term resembling an inertial force (proportional to the velocity) in the equation for u_x. The effect of this term is to cause the shifts in the eigenvalues and, thereby, to introduce a strong angular dependence in the Green's function. This term is also responsible for breaking the degeneracy of the shear modes. The second effect of the flow field is the introduction of the linear mode coupling which also contributes to the spatial asymmetry of the Green's function through its effect on the eigenvectors. In addition to this "static" effect, the linear mode coupling strongly modifies the temporal evolution of fluctuations and, in fact, is necessary to understand the stability of the modes. These hydrodynamic effects have obvious relevance to the evolution of fluctuations which can be calculated from the Green's function given in Eq. (3.19). As an example, the evolution of a sound excitation has been exhibited and the expected kinematic effects (i.e., change of frequency) due to convection are evident.

These purely hydrodynamic modifications of the dynamics of fluctuations in the nonequilibrium state also characterize the statistical mechanics (ensemble) for this state, through the fluctuation-dissipation relations, (4.9) or (4.11). For example, the equal-time correlation functions completely determine the covariance for the steady-state distribution function. The most striking feature of the correlation functions is that they are long ranged. Although this had been recognized from small-shear-rate calculations, the analysis here gives the detailed form of this space dependence at large shear rates as well. The one-dimensional correlation of densities in Fig. 2 has an exact $x^{-5/3}$ power law dependence, for large distances. This suggests that the angle-averaged radial distribution function decays as $r^{-11/3}$ for large r, in contrast to the r^{-1} behavior for small shear rates. The light scattering function (4.17) reflects both the dynamical and statistical mechanical changes due to shear flow. The qualitative changes in shape of the Brillouin and Rayleigh lines are a direct consequence on the nonequilibrium Onsager regression law (4.10). The wave-vector and shear-rate dependence of the Landau-Placzek ratio is a direct measure of the nonequilibrium fluctuation-dissipation relation (4.11). Unfortunately, all of these effects are too small to be accurately measured by conventional Brillouin scattering techniques. However, with a suitable modification of the Langevin model to apply at larger wave vectors, related predictions could be amenable to test by computer simulation.

As noted in the Introduction, many intersting and unexplained phenomena are expected to be due to nonlinear coupling of the hydrodynamic modes described here. Such coupling occurs as an integration over all wave vectors $\mathbf{k}$, with the asymptotically small k values domination. Consequently, even though some of the conditions considered here (e.g., $a/\Gamma_0 k^2=1$) cannot be attained simply in the laboratory, they do play a key role in the calculations of nonlinear mode coupling. As an application of the results obtained here, we have calculated the lowest-order mode-coupling contributions to the nonlinear shear viscosity $\eta(a)$ as defined in Eq. (1.1). Equation (3.3), which is a macroscopic equation, has been used to define the renormalized shear viscosity. By averaging the microscopic equation for energy conservation, the second of Eqs. (2.2), and comparing the result to Eq. (3.3) we obtain an expression for $\eta(a)$ in terms of integrals of correlation functions.[39] The result is Eq. (1.1) with

$$\eta_1 = -10^{-3} k_B T_0 \left[\frac{2.56}{(2D_{0T})^{3/2}} + \frac{4.17}{(\Gamma_0)^{3/2}} \right] ,$$

in agreement with Ernst *et al.*[2,40] Furthermore, it can be shown that the analytic expressions obtained for η_1 (for an incompressible fluid) by Yamada and Kawasaki agree with ours, the differences in the value of n_1 being attributable to errors in the numerical analysis. A more complete analysis of the fluctuation renormalization of the macroscopic equations in both two and three dimensions using the hydrodynamic modes calculated here is in progress.

ACKNOWLEDGMENTS

The authors wish to thank Bob Coldwell for assistance with the numerical analysis. One of the authors (J.L.) wishes to thank David Kilcrease and Pradeep Kumar for several useful discussions. This research was supported by National Science Foundation Grant No. CHE-84-11932.

APPENDIX A: NONLINEAR NAVIER-STOKES-LANGEVIN EQUATIONS

The microscopic conservation laws in the form of Eqs. (2.2) and (2.3) are

$$\left[\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla \right] \rho + \rho \nabla\cdot\mathbf{v} = 0 ,$$

$$\left[\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla \right] u' + h\nabla\cdot\mathbf{u}' + \nabla\cdot\mathbf{s}^* + t_{ij}^* \frac{\partial v_i}{\partial r_j} = -\left[\nabla\cdot\mathbf{s}^R + t_{ij}^R \frac{\partial v_i}{\partial r_j} \right] , \quad \text{(A1)}$$

$$\rho \left[\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla \right] v_i + \frac{\partial}{\partial r_j}(p\delta_{ij} + t_{ij}^*) = -\frac{\partial}{\partial r_j} t_{ij}^R .$$

The irreversible fluxes $\mathbf{s}^*$ and t_{ij}^* are given by Eqs. (2.5) in the Navier-Stokes limit. The random components of the fluxes are defined in terms of three dimensionless stochastic variables,[30]

$$s_i^R(\mathbf{r},t) = (T^2\lambda_B)^{1/2} f_i^{(1)} ,$$
$$t_{ij}^R(\mathbf{r},t) = (T\eta_B)^{1/2} f_{ij}^{(2)} + (T\kappa_B)^{1/2} f_{ij}^{(3)} . \quad \text{(A2)}$$

Here λ_B, η_B, and κ_B are the bare transport coefficients, and T is the microscopic temperature defined in Eq. (2.6). The $\{f^{(\alpha)}(\mathbf{r},t)\}$ are a set of independent random variables characterizing a Gaussian-Markovian process, with mean values and covariances,

$$\langle f^{(\alpha)} \rangle = 0, \quad \text{all } \alpha$$
$$\langle f^{(\alpha)} f^{(\beta)} \rangle = 0, \quad \alpha \neq \beta$$
$$\langle f_i^{(1)}(\mathbf{r},t) f_j^{(1)}(\mathbf{r}',t') \rangle = 2\delta_{ij}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t') , \quad \text{(A3)}$$
$$\langle f_{ij}^{(2)}(\mathbf{r},t) f_{kl}^{(2)}(\mathbf{r}',t') \rangle = 2\Delta_{ijkl}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t') ,$$
$$\langle f_{ij}^{(3)}(\mathbf{r},t) f_{kl}^{(3)}(\mathbf{r}',t') \rangle = 2\delta_{ij}\delta_{kl}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t') .$$

The tensor Δ_{ijkl} is defined by

$$\Delta_{ijkl} = \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \tfrac{2}{3}\delta_{ij}\delta_{kl} . \quad \text{(A4)}$$

Equations (A1)–(A3) provide a closed description for the macroscopic dynamics and fluctuations in a simple fluid.

The macroscopic hydrodynamic equations have the same form as (A1), without the random components of the fluxes. However, the irreversible part of the fluxes will generally differ from the average of Eqs. (2.3). This is due to fluctuation renormalization by the nonlinear terms in the Langevin equation. The latter present technical difficulties associated with differences between the average of a function of the microscopic variables and the corresponding function of their averages. These complications make the detailed relationship of the macroscopic hydrodynamic equations to the Langevin equations somewhat indirect. To be more specific, if the macroscopic mass, energy, and momentum densities are distinguished from their microscopic counterparts by a subscript, 0, the two are related by the definitions

$$\rho_0 \equiv \langle \rho \rangle, \quad u_0 \equiv \langle u \rangle, \quad \mathbf{p}_0 \equiv \langle \mathbf{p} \rangle . \quad \text{(A5)}$$

The macroscopic flow velocity $\mathbf{v}_0$, internal energy ε_0, pressure p_0, and temperature T_0 are defined by

$$\mathbf{p}_0 = \rho_0 \mathbf{v}_0, \quad \varepsilon_0 = u_0 - \tfrac{1}{2}\rho_0 v_0^2 ,$$
$$p_0 = p_e(\rho_0, \varepsilon_0), \quad T_0 = T_e(\rho_0, \varepsilon_0) . \quad \text{(A6)}$$

The quantities in (A6) are closely related to, but not equal to, the average of the corresponding variables in the Langevin equation. For example, the average microscopic velocity and internal energy are related to $\mathbf{v}_0$ and ε_0,

$$\langle \rho \mathbf{v} \rangle = \rho_0 \mathbf{v}_0 ,$$
$$\langle u' \rangle = \varepsilon_0 + \tfrac{1}{2}(\rho_0 v_0^2 - \langle \rho v^2 \rangle) . \quad \text{(A7)}$$

Similarly, $p_e(\rho_0,\varepsilon_0)$ and $T_e(\rho_0,\varepsilon_0)$ differ from the averages of $p_e(\rho,u')$ and $T_e(\rho,u')$ due to their nonlinear dependence on the arguments of these functions.

The linearization of Eqs. (A1) around uniform shear flow, defined by Eqs. (3.1) and (3.3), gives

$$\left[\frac{\partial}{\partial t} + a_{ij} r_j \frac{\partial}{\partial r_i} \right] \delta\rho + \rho_0 \nabla\cdot\delta\mathbf{v} = 0 ,$$

$$\left[\frac{\partial}{\partial t} + a_{ij} r_j \frac{\partial}{\partial r_i} \right] \delta\varepsilon + h_0 \nabla\cdot\delta\mathbf{v} - \lambda_B \left[\left[\frac{\partial T_0}{\partial \varepsilon_0} \right]_{\rho_0} \nabla^2 \delta\varepsilon + \left[\frac{\partial T_0}{\partial \rho_0} \right]_{\varepsilon_0} \nabla^2 \delta\rho \right] - 2\eta_B a_{ij} \left[\frac{\partial}{\partial r_j}\delta v_i + \frac{\partial}{\partial r_i}\delta v_j \right] = -(\nabla\cdot\mathbf{s}^R + a_{ij} t_{ij}^R) , \quad \text{(A8)}$$

$$\left[\frac{\partial}{\partial t}+a_{ij}r_j\frac{\partial}{\partial r_i}\right]\delta v_i+a_{ij}\delta v_j$$
$$+\rho_0^{-1}\left[\left[\frac{\partial p_0}{\partial \varepsilon_0}\right]_{\rho_0}\frac{\partial}{\partial r_i}\delta\varepsilon+\left[\frac{\partial p_0}{\partial \rho_0}\right]_{\varepsilon_0}\frac{\partial}{\partial r_i}\delta\rho\right]$$
$$+\nu_B\nabla^2\delta v_i-(\tfrac{1}{3}\nu_B+\rho_0^{-1}\kappa_B)\frac{\partial}{\partial r_i}\nabla\cdot\delta\mathbf{v}=-\rho_0^{-1}\frac{\partial}{\partial r_j}t_{ij}^R .$$

Here $\nu_B\equiv\rho_0^{-1}\eta_B$, and all bare transport coefficients are now functions of ρ_0 and ε_0. Also, the fluctuations are defined by

$$\delta\rho\equiv\rho-\langle\rho\rangle,\quad \delta\varepsilon\equiv u'-\langle u'\rangle,\quad \delta\mathbf{v}\equiv\mathbf{v}-\langle\mathbf{v}\rangle . \tag{A9}$$

The Fourier transform [Eq. (2.12)] of Eqs. (A8) has the general form

$$\left[\frac{\partial}{\partial t}-a_{ij}k_i\frac{\partial}{\partial k_j}\right]\tilde{z}_\alpha(\mathbf{k},t)+L_{\alpha\beta}(\mathbf{k},a,t)\tilde{z}_\beta(\mathbf{k},t)=\overline{R}_\alpha(\mathbf{k},t) . \tag{A10}$$

The matrix $L_{\alpha\beta}(\mathbf{k},a,t)$ is given by

$$L_{\alpha\beta}(\mathbf{k},a,t)=-ikB_{\alpha\beta}(t)+k^2C_{\alpha\beta}(t)+aD_{\alpha\beta}(\hat{\mathbf{k}},t)+a^2E_{\alpha\beta}(t) , \tag{A11}$$

where $\hat{\mathbf{k}}$ is a unit vector,

$$B_{\alpha\beta}=\begin{bmatrix}0&0&\rho_0c_1&0&0\\0&0&h_0c_2&0&0\\\rho_0c_1&h_0c_2&0&0&0\\0&0&0&0&0\\0&0&0&0&0\end{bmatrix},$$

$$C_{\alpha\beta}=\begin{bmatrix}0&0&0&0&0\\c_3\lambda_0&c_4\lambda_0&0&0&0\\0&0&\nu_0'&0&0\\0&0&0&\nu_0&0\\0&0&0&0&\nu_0\end{bmatrix}, \tag{A12}$$

$$D_{\alpha\beta}=\begin{bmatrix}0&0&0&0&0\\0&0&0&0&0\\0&0&\Gamma_{11}&\Gamma_{12}&\Gamma_{13}\\0&0&\Gamma_{21}&\Gamma_{22}&\Gamma_{23}\\0&0&\Gamma_{31}&\Gamma_{32}&\Gamma_{33}\end{bmatrix},$$

$$a^2E_{\alpha\beta}(t)=\delta_{\alpha1}\delta_{\beta1}\left[\frac{\partial \ln c_1}{\partial\varepsilon_0}\right]_{\rho_0}+\delta_{\alpha2}\delta_{\beta2}\left[\frac{\partial \ln c_2}{\partial\varepsilon_0}\right]_{\rho_0},$$

and Γ_{ij} is given by

$$a\Gamma_{ij}=\hat{e}_l^{(i)}(\mathbf{k})a_{lm}\hat{e}_m^{(j)}(\mathbf{k})-a_{lm}k_l\hat{e}_n^{(i)}(\mathbf{k})\frac{\partial}{\partial k_m}\hat{e}_n^{(j)}(\mathbf{k}) . \tag{A13}$$

The parameters c_i are given by

$$\begin{aligned}
c_0^2&=(\rho_0c_1)^2+(h_0c_2)^2=\left[\frac{\partial p_0}{\partial\rho_0}\right]_{s_0},\\
c_1^2&=\rho_0^{-2}\left[\frac{\partial p_0}{\partial\rho_0}\right]_{\varepsilon_0}=\frac{c_0^2}{\rho_0^2}\left[1-\frac{\alpha_T}{\rho_0c_p}h_0\right],\\
c_2^2&=(\rho_0h_0)^{-1}\left[\frac{\partial p_0}{\partial\varepsilon_0}\right]_{\rho_0}=\frac{c_0^2\alpha_T}{\rho_0h_0c_p},\\
c_3&=\frac{c_2}{c_1}\left[\frac{\partial T_0}{\partial\rho_0}\right]_{\varepsilon_0}=\frac{c_2}{c_1}\left[\frac{\gamma-1}{\rho_0\alpha_T}-\frac{h_0}{\rho_0c_v}\right],\\
c_4&=\left[\frac{\partial T_0}{\partial\varepsilon_0}\right]_{\rho_0}=\frac{1}{\rho_0c_v} .
\end{aligned} \tag{A14}$$

Also, $\nu_0=\eta_B/\rho_0$ is the kinematic viscosity, $\nu_0'=\frac{4}{3}\nu_0+\rho_0^{-1}\kappa_B$, $\alpha_T=\rho_0^{-1}(\partial\rho_0/\partial T_0)|_p$ is the thermal expansion coefficient, c_v is the constant volume specific heat, c_p is the constant pressure specific heat, $\gamma=c_p/c_v$, and c_0 is the speed of sound.

Finally, the random forces $\overline{R}_\alpha$ are

$$\begin{aligned}
\overline{R}_\alpha&\equiv R_\alpha-\langle R_\alpha\rangle ,\\
R_1&=0 ,\\
R_2&=c_1\left[i\mathbf{k}\cdot\tilde{\mathbf{s}}^R(\mathbf{k},t)-a_{ij}\tilde{t}_{ij}^R(\mathbf{k},t)\right] ,\\
R_{2+l}&=\hat{\mathbf{e}}_i^{(l)}(\mathbf{k})ik_j\tilde{t}_{ij}^R(\mathbf{k},t)/\rho,\quad l=1,2,3 .
\end{aligned} \tag{A15}$$

The effect of the shear flow occurs in several terms of (A10). First, the linear mode-coupling term (i.e., the gradient term in $\mathbf{k}$) is proportional to the shear rate and vanishes in equilibrium. The contribution from $E_{\alpha\beta}$ is proportional to a^2 and is due to the viscous heating expressed by Eq. (3.3). The time dependence of $L_{\alpha\beta}(\mathbf{k},t)$ is also due to this heating and occurs only through the dependence on $\varepsilon_0(t)$. Aside from the neglect of terms nonlinear in $z_\alpha(\mathbf{k},t)$, no other approximations have been made. In particular, there has been no restriction that the macroscopic state of uniform shear be close to equilibrium.

The tensor Γ_{ij} is considerably simplified by a suitable choice for the unit vectors $\hat{\mathbf{e}}^{(\alpha)}$. In particular, we choose $\hat{\mathbf{e}}^{(1)}=\mathbf{k}/k$, $\hat{\mathbf{e}}^{(2)}$ perpendicular to $\hat{\mathbf{e}}^{(1)}$ and in the $\mathbf{k}$-$\hat{\mathbf{y}}$ plane, and $\hat{\mathbf{e}}^{(3)}$ perpendicular to $\hat{\mathbf{e}}^{(1)}$ and $\hat{\mathbf{e}}^{(2)}$,

$$\begin{aligned}
\hat{\mathbf{e}}^{(1)}&=\hat{\mathbf{k}} ,\\
\hat{\mathbf{e}}^{(2)}&=[\hat{\mathbf{y}}-\hat{\mathbf{e}}^{(1)}(\hat{\mathbf{e}}^{(1)}\cdot\hat{\mathbf{y}})]/\hat{k}_\perp ,\\
\hat{\mathbf{e}}^{(3)}&=\hat{\mathbf{e}}^{(1)}\times\hat{\mathbf{e}}^{(2)} ,
\end{aligned} \tag{A16}$$

where $\hat{k}_\perp=(k^2-k_y^2)^{1/2}/k$. It then follows that

$$\Gamma_{11}=-\Gamma_{22}=k_xk_y/k^2\ ,$$
$$\Gamma_{12}=-k_x/k_\perp\ ,$$
$$\Gamma_{31}=-k_yk_z/kk_\perp\ , \tag{A17}$$
$$\Gamma_{32}=-k_z/k\ ,$$
$$\Gamma_{ij}=0,\ \text{all others}\ .$$

APPENDIX B: HYDRODYNAMIC MODES

The general solution to the linear hydrodynamic equations may be determined from the generalized eigenvalue problem, Eq. (3.10):

$$\left[-a_{ij}k_i\frac{\partial}{\partial k_j}+L\right]\zeta^{(i)}=\lambda_i\zeta^{(i)}\ . \tag{B1}$$

The differential operator with respect to k makes this a nonlinear eigenvalue equation. The problem is posed in a five-dimensional complex Hilbert space with scalar product

$$(a,b)=\sum_\alpha a_\alpha^* b_\alpha\ . \tag{B2}$$

Since L is not Hermitian, the eigenvectors will not be pairwise orthogonal in general. The associated biorthogonal set is denoted by $\{\eta^{(i)}\}$,

$$(\eta^{(i)},\zeta^{(i)})=\delta_{ij}\ . \tag{B3}$$

The perturbation theory is generated by the expansions

$$\lambda_i=k\lambda_{i,0}+k^2\lambda_{i,1}+\cdots\ ,$$
$$\zeta^{(i)}=\zeta_0^{(i)}+k\zeta_1^{(i)}+\cdots\ . \tag{B4}$$

Substitution into Eq. (B1) and use of the form (A11) for L gives the equations for first- and second-order perturbation theory,

$$(-iB-\lambda_{i,0}I)\zeta_0^{(i)}=0\ ,$$
$$(-iB-\lambda_{i,0}I)\zeta_1^{(i)}=\left[\lambda_{i,1}I-C-ak^{-2}D+k^2a_{ij}k_i\frac{\partial}{\partial k_j}\right]\zeta_0^{(i)}\ , \tag{B5}$$

where I denotes the identity. The eigenvalues of the matrix $-iB$ are readily found from (A12) to be

$$\lambda_{1,0}=-ic_0,\quad \lambda_{2,0}=+ic_0\ ,$$
$$\lambda_{3,0}=\lambda_{4,0}=\lambda_{5,0}=0\ , \tag{B6}$$

and the corresponding eigenvectors are

$$\psi^{(1)}=\frac{1}{\sqrt{2}c_0}(\rho_0c_1,h_0c_2,c_0,0,0)\ ,$$
$$\psi^{(2)}=\frac{1}{\sqrt{2}c_0}(\rho_0c_1,h_0c_2,-c_0,0,0)\ ,$$
$$\psi^{(3)}=\frac{1}{c_0}(h_0c_2,-\rho_0c_1,0,0,0)\ , \tag{B7}$$
$$\psi^{(4)}=(0,0,0,1,0)\ ,$$
$$\psi^{(5)}=(0,0,0,0,1)\ .$$

These vectors also form an orthonormal basis. Because of the degeneracy for $i=3,4,5$, the lowest-order eigenvectors $\{\zeta_0^{(i)}\}$ are

$$\zeta_0^{(1)}=\psi^{(1)}\ ,$$
$$\zeta_0^{(2)}=\psi^{(2)}\ , \tag{B8}$$
$$\zeta_0^{(i)}=\sum_{j=3}^{5}M_{ij}\psi^{(j)},\quad i=3,4,5\ .$$

The eigenvalues to next-order perturbation theory and the coefficients M_{ij} are determined from the second of Eqs. (B5). Taking the scalar product of this equation with $\psi^{(i)}$ and use of (B8) gives

$$\lambda_{i,1}=[\psi^{(i)},(C+ak^{-2}D)\psi^{(j)}],\quad i=1,2$$
$$\sum_{l=3}^{5}\left[[\psi^{(j)},(C+ak^{-2}D)\psi^{(l)}]-\left[k^{-2}a_{ij}k_i\frac{\partial}{k_j}+\lambda_{i,1}\right]\delta_{jl}\right]M_{il}=0,\quad i,j=3,4,5\ . \tag{B9}$$

The specific forms of the matrices C and D are displayed in Eqs. (A12). It follows immediately that for $i=1,2$,

$$\lambda_{i,1}=\tfrac{1}{2}(\Gamma_0+ak^{-2}\Gamma_{11}),\quad i=1,2\ . \tag{B10}$$

Here

$$\Gamma_0=\frac{h_0\lambda_0c_2}{c^2}(\rho_0c_1c_3+h_0c_2c_4)+\nu_0'=\frac{c_p}{c_v}D_T+\nu_0'$$

is the sound damping constant, D_T is the thermal diffusivity, and Γ_{11} is defined by Eq. (A13). Also, it is seen from the form of C and D that for $l=3,4,5$,

$$[\psi^{(3)},(C+ak^{-2}D)\psi^{(l)}]=\delta_{3,l}D_T\ . \tag{B11}$$

Consequently, a solution for $i=3$ is

$$\lambda_{3,1}=D_T\ ,$$
$$M_{3l}=\delta_{3l}\ . \tag{B12}$$

The remaining two eigenvalues are determined from (B9), which now has the simple form, for $i,j=4,5$,

$$\sum_{l=4,5}\left[ak^{-2}(\psi^{(j)},D\psi^{(l)})-\left[(\lambda_{i,1}-\nu_0)+k^{-2}a_{lm}k_l\frac{\partial}{\partial k_m}\right]\delta_{jl}\right]M_{il}=0\ . \tag{B13}$$

The elements of $(\psi^{(j)},D\psi^{(l)})$ are found to be

$$(\psi^{(j)},D\psi^{(l)})=\begin{bmatrix}\Gamma_{22} & 0\\ \Gamma_{32} & 0\end{bmatrix}\ , \tag{B14}$$

where the special coordinate system (A16) has now been selected for simplicity. Equations (B13) are then, for $i=4,5$,

$$\left[\lambda_{i,1}-\nu_0-(a/k^2)\Gamma_{22}+(a/k^2)k_x\frac{\partial}{\partial k_y}\right]M_{i4}=0\ ,$$
$$\left[\lambda_{i,1}-\nu_0+(a/k^2)k_x\frac{\partial}{\partial k_y}\right]M_{i5}-(a/k^2)\Gamma_{32}M_{i4}=0\ . \tag{B15}$$

Solutions to these equations include

$$\lambda_{4,1}=\nu_0+ak^{-2}\Gamma_{22},\quad \lambda_{5,1}=\nu_0 \tag{B16}$$

with the coefficients

$$M_{44}=1,\quad M_{45}=M\ ,$$
$$M_{54}=0,\quad M_{55}=1\ ,$$
$$M\equiv-\frac{kk_z}{k_xk_\perp}\tan^{-1}(k_y/k_\perp)\ ,$$
$$k_\perp^2=k^2-k_y^2\ . \tag{B17}$$

These results clearly apply only if $k_x\neq 0$. More general solutions to (B15) exist with different eigenvalues and coefficients. However, it is possible to show that the Green's functions defined by Eqs. (3.14) and (3.16) are invariant with respect to these differences. More specifically, any differences in the eigenvalues cancel in the Green's function so that only the hydrodynamic modes determined by the choice made here appears in the final results.

In summary, the eigenvalues and eigenvectors to this order are

$$\lambda_1=-ick+\tfrac{1}{2}(\Gamma k^2+ak_xk_y/k^2)\ ,$$
$$\lambda_2=\lambda_1^*\ ,$$
$$\lambda_3=D_Tk^2\ ,$$
$$\lambda_4=\nu_0k^2-ak_xk_y/k^2\ ,$$
$$\lambda_5=\nu_0k^2\ , \tag{B18}$$

and the biorthogonal set to lowest order is found to be

$$\zeta^{(i)}=\eta^{(i)}=\psi^{(i)},\quad i=1,2,3$$
$$\zeta^{(4)}=(0,0,0,1,M)\ ,$$
$$\zeta^{(5)}=(0,0,0,0,1)\ ,$$
$$\eta^{(4)}=(0,0,0,1,0)\ ,$$
$$\eta^{(5)}=(0,0,0,-M,1)\ . \tag{B19}$$

APPENDIX C: CORRELATION FUNCTIONS

The autocorrelation matrix for the random forces is given by Eq. (4.6),

$$\langle F^{(i)}(\mathbf{k},t)F^{(j)}(\mathbf{k}',t')\rangle\equiv(2\pi)^3\delta(\mathbf{k}+\mathbf{k}')\delta(t-t')F^{(ij)}(\mathbf{k})\ , \tag{C1}$$

where $F^{(i)}(\mathbf{k},t)$ is defined by

$$F^{(i)}(\mathbf{k},t)\equiv\eta_\alpha^{(i)}(\mathbf{k})\bar{R}_\alpha(\mathbf{k},t) \tag{C2}$$

and the random forces are given in Eqs. (A15). The forces $R_\alpha(\mathbf{k},t)$ are linear combinations of the random parts of the heat flux and pressure tensor of Eqs. (A2) and (A3). To order k^2, it is found that

$$\langle s_i^R(\mathbf{k},t)\rangle=0+O(k^2)\ ,$$
$$\langle t_{ij}^R(\mathbf{k},t)\rangle=0+O(k^2)\ ,$$
$$\langle s_i^R(\mathbf{k},t)t_{jl}^R(\mathbf{k}',t')\rangle=0\ ,$$
$$k_ik_j'\langle s_i^R(\mathbf{k},t)s_j^R(\mathbf{k}',t')\rangle=-(2\pi)^3\delta(\mathbf{k}+\mathbf{k}')\delta(t-t')2k^2T_0^2\lambda_0\ ,$$
$$k_ik_j'e_m^{(l)}(\mathbf{k})e_n^{(P)}(\mathbf{k}')\langle t_{im}^R(\mathbf{k},t)t_{jn}^R(\mathbf{k}',t')\rangle=(2\pi)^3\delta(\mathbf{k}+\mathbf{k}')\delta(t-t')\times 2T_0k^2[2\eta_B\delta_{l1}\delta_{P1}+\delta_{lP}(\kappa_B-\tfrac{2}{3}\eta_B)]\ . \tag{C3}$$

The matrix of forces in (C1) then becomes

$$F^{(ij)}(\mathbf{k})=2k^2T_0\sum_{\alpha=1}^{5}R_\alpha\eta_\alpha^{(i)}(\mathbf{k})\eta_\alpha^{(j)}(-\mathbf{k}) \tag{C4}$$

with

$$R_1=0,\quad R_2=c_2^2\lambda_BT_0^2,\quad R_3=-\nu_0'T_0\rho_0,$$
$$R_4=-R_5=\nu_0T_0\rho_0\ . \tag{C5}$$

Evaluating the sum in (C4) gives the desired results

$$F^{(ij)}(k)=F^{(ji)}(k)\ \text{ for } i,j=1,2,3\ ,$$
$$F^{14}=F^{15}=F^{24}=F^{25}=F^{34}=F^{35}=0\ ,$$
$$F^{11}=F^{22}=k^2T_0(\Gamma_0-2\nu_0')\ ,$$
$$F^{12}=k^2T_0\Gamma_0\ ,$$
$$F^{13}=F^{23}=-k^2\sqrt{2}\lambda_0\rho_0h_0c_1c_2\left[\frac{T_0c_2}{c_0}\right]^2\ ,$$
$$F^{33}=-2k^2\nu_0'T_0\left[\frac{\rho_0c_2}{c_0}\right]^2=-2k^2\nu_0'T_0\frac{c_0^2\alpha_T}{h_0c_p}\rho_0\ ,$$
$$F^{44}=2k^2T_0\nu_0/\rho_0\ ,$$
$$F^{45}=-F^{54}=M(\mathbf{k})F^{44}\ ,$$
$$F^{55}=-[1+M^2(\mathbf{k})]F^{44}\ . \tag{C6}$$

The equal-time correlation functions in Sec. IV are determined from the functions $C_{\alpha\beta}(\mathbf{k};a)$ of Eqs. (4.8) and (4.9),

$$C_{\alpha\beta}(\mathbf{k};a)=\int_0^\infty d\tau\sum_{i,j}\xi_\alpha^{(i)}(\mathbf{k},\tau)\xi_\beta^{(j)}(-\mathbf{k},\tau)F^{(ij)}(k(-\tau))$$
$$=\int_0^\infty d\tau\sum_{i,j}E^{(i)}(\mathbf{k},\tau)E^{(j)}(-\mathbf{k},\tau)\xi_\alpha^{(i)}(\mathbf{k})\xi_\beta^{(j)}(-\mathbf{k})F^{(ij)}(\mathbf{k}(-\tau))\ . \tag{C7}$$

At this point, it is convenient to convert the variables $\tilde{z}_1$ and $\tilde{z}_2$ back to $\delta\tilde{\rho}$ and $\delta\tilde{\varepsilon}$ using their definitions (3.5), e.g.,

$$C_{\rho\rho}(\mathbf{k};a)=c_1^{-2}C_{11}(\mathbf{k};a) ,$$

etc. The correlation functions can then be written as

$$\begin{aligned}
C_{\rho\rho}(\mathbf{k};a)&=k_BT_0\rho_0^2\chi_T[1+\Delta_1(\mathbf{k};a)] ,\\
C_{\rho\varepsilon}(\mathbf{k};a)&=k_BT_0h_0\rho_0\chi_T\left[1-h_0^{-1}T_0\frac{\alpha_T}{\chi_T}+\Delta_1(\mathbf{k};a)\right] ,\\
C_{\varepsilon\varepsilon}(\mathbf{k};a)&=k_BT_0\chi_T\left[\chi_T^{-1}T_0c_v+\left[h_0-T_0\frac{\alpha_T}{\chi_T}\right]^2+h_0^2\Delta_1(\mathbf{k};a)\right] ,\\
C_{33}(\mathbf{k};a)&=k_BT_0\rho_0^{-1}[1+\gamma\Delta_1(\mathbf{k};a)] ,\\
C_{44}(\mathbf{k};a)&=k_BT_0\rho_0^{-1}[1-\Delta_2(\mathbf{k};a)] ,\\
C_{45}(\mathbf{k};a)&=k_BT_0\rho_0^{-1}\Delta_3(\mathbf{k};a) ,\\
C_{55}(\mathbf{k};a)&=k_BT_0\rho_0^{-1}[1-\Delta_4(\mathbf{k};a)] .
\end{aligned} \tag{C8}$$

The functions $\Delta_i(\mathbf{k};a)$ all go to zero with the shear rate and represent the corrections to the local equilibrium results. Explicitly,

$$\begin{aligned}
\Delta_1(\mathbf{k};a)&=\gamma^{-1}a\int_0^\infty dt\frac{kk_xk_y(-t)}{k^3(-t)}e^{-\Gamma_0k^2\beta(-t)} ,\\
\Delta_2(\mathbf{k};a)&=2a\int_0^\infty dt\frac{k_xk_y(-t)}{k^2}e^{-2\nu_0k^2\beta(-t)} ,\\
\Delta_3(\mathbf{k};a)&=a\int_0^\infty dt\left[\frac{2k_xk_y(-t)}{kk(-t)}F(\mathbf{k},t)+\frac{k_z}{k}\right]\times e^{-2\nu_0k^2\beta(-t)} ,\\
\Delta_4(\mathbf{k};a)&=2a\int_0^\infty dt\,F(\mathbf{k},t)\left[\frac{k_xk_y(-t)}{k^2(-t)}F(\mathbf{k},t)+\frac{k_z}{k}\right]\times e^{-2\nu_0k^2\beta(-t)} ,
\end{aligned} \tag{C9}$$

where

$$F(\mathbf{k},t)=M(\mathbf{k}(-t))-\frac{k(-t)}{k}M(\mathbf{k}) . \tag{C10}$$

The light scattering function is defined by Eq. (4.17). Representation of the density autocorrelation function in terms of the hydrodynamic modes leads to the form [using Eqs. (4.10) and (4.7)]

$$\begin{aligned}
S(\mathbf{k};\omega)&=\sum_{i=1}^{3}S^{(i)}(\mathbf{k};\omega) ,\\
S^{(i)}(\mathbf{k};\omega)&\equiv 2R_e\int\frac{d\mathbf{k}_1}{(2\pi)^3}\xi_1^{(i)}(\mathbf{k}_1)\tilde{\theta}(\mathbf{k}-\mathbf{k}_1)\bar{\sigma}^{(i)}(\mathbf{k}_1;\omega) ,
\end{aligned} \tag{C11}$$

where R_e denotes the real part, and $\bar{\sigma}^{(i)}(\mathbf{k}_1;\omega)$ is the solution to

$$\left[-i\omega+\lambda_i(\mathbf{k}_1,a)-a_{ij}k_{1i}\frac{\partial}{\partial k_{1j}}\right]\bar{\sigma}^{(i)}(\mathbf{k}_1;\omega)=\tilde{\theta}(\mathbf{k}-\mathbf{k}_1)\eta_\alpha^{(i)}(k_1)c_1^{-2}C_{\alpha 1}(\mathbf{k}_1;a) . \tag{C12}$$

The index i in (C11) and (C12) only ranges over 1—3 because the density does not couple to the transverse modes. Further simplification of these results is possible by noting that to the order in perturbation theory considered here $\xi_1^{(i)}(k)$ is independent of k for $i=1$—3. Then, multiplying (4.19) by $\xi_1^{(i)}\tilde{\theta}(\mathbf{k}-\mathbf{k}_1)$ and integrating over k_1 gives

$$\int\frac{d\mathbf{k}_1}{(2\pi)^3}\left[\tilde{\theta}^2(\mathbf{k}-\mathbf{k}_1)\{[-i\omega+\lambda_i(\mathbf{k}_1,a)]\sigma(\mathbf{k}_1;\omega)-\xi_1^{(i)}\eta_\alpha^{(i)}c_1^{-2}C_{\alpha 1}(\mathbf{k}_1;a)\}-\tilde{\theta}(k-k_1)a_{ij}k_{1i}\frac{\partial}{\partial k_{1j}}\tilde{\theta}(\mathbf{k}-\mathbf{k}_1)\sigma(\mathbf{k}_1;\omega)\right]=0 , \tag{C13}$$

where $\xi_1(i)\bar{\sigma}^{(i)}(k_1;\omega)\equiv\tilde{\theta}(\mathbf{k}-\mathbf{k}_1)\sigma^{(i)}(k_1;\omega)$. The last term in Eq. (C13) can be transformed by partial integration to yield

$$\int\frac{d\mathbf{k}_1}{(2\pi)^3}\tilde{\theta}^2(\mathbf{k}-\mathbf{k}_1)\left[\left[-i\omega+\lambda_i(\mathbf{k}_1,a)-\tfrac{1}{2}a_{ij}k_{1i}\frac{\partial}{\partial k_{1j}}\right]\sigma^{(i)}(\mathbf{k}_1;\omega)-\xi_1^{(i)}\eta_\alpha^{(i)}c_1^{-2}C_{\alpha 1}(\mathbf{k}_1;a)\right]=0 . \tag{C14}$$

Equations (C11) may also be written in terms of $\sigma^{(i)}(k_1;\omega)$,

$$S^{(i)}(\mathbf{k};\omega)=2R_e\int\frac{d\mathbf{k}_1}{(2\pi)^3}\tilde{\theta}^2(k_1)\sigma^{(i)}(\mathbf{k}+\mathbf{k}_1;\omega) . \tag{C15}$$

Typically, the dimensions of the light scattering region can be large compared to the change in wavelength, so that $k_1 \ll k$. Then the functions $S^{(i)}(k;\omega)$ can be written

$$S^{(i)}(\mathbf{k};\omega)=2R_e\sigma^{(i)}(\mathbf{k};\omega) , \tag{C16}$$

where, with Eq. (C14), the $\sigma^{(i)}(\mathbf{k};\omega)$ satisfy the approximate equations

$$\left[-i\omega+\lambda_i(\mathbf{k},a)-\tfrac{1}{2}a_{ij}k_i\frac{\partial}{\partial k_j}\right]\sigma^{(i)}(\mathbf{k};\omega)=c_1^{-2}\xi_1^{(i)}\eta_\alpha^{(i)}C_{\alpha 1}(\mathbf{k};a) , \tag{C17}$$

and the integral over $\tilde{\theta}^2(k)$ has been arbitrarily set equal to unity. The right side of (C17) is the same for $i=1$ and 2, corresponding to the two sound modes so that $S^{(1)}(\mathbf{k},\omega)=S^2(\mathbf{k},-\omega)\equiv S_B(\mathbf{k},\omega)$. The third contribution is from the heat mode. In summary, the light scattering function is

$$S(\mathbf{k},\omega)=S_B(\mathbf{k},\omega)+S_B(\mathbf{k},-\omega)+S_R(\mathbf{k},\omega) . \tag{C18}$$

The first two terms give the Brillouin peaks and the last gives the Rayleigh peak.

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[40]Note that the sound damping constant Γ_0 as defined here is twice that of Ernst *et al.*, Ref. 2.