Bilinear Contributions to Equilibrium Correlation Functions^{*}

T. Keyes[†] and Irwin Oppenheim

Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 20 November 1972)

The long-time behavior of autocorrelation functions of conserved variables and of their corresponding dissipative fluxes is studied using the Mori formalism. A set of variables including linear and bilinear forms of the conserved variables is treated and consistent expansions to quadratic order in the wave-vector dependence of the appropriate correlation functions are carried out. The treatment is self-consistent and does not involve factorization of time-dependent correlation functions. Explicit calculations for the wave vector \vec{k} and the frequency-dependent shear-viscosity coefficient are carried out in two dimensions. The asymptotic time dependence of the shear-viscosity correlation function is proportional to t^{-1} when $k \rightarrow 0$. The effect of higher-order nonlinear forms of the conserved variables has not been studied; such a study is essential to ensure the validity of the asymptotic time dependence found here.

I. INTRODUCTION

There has been much recent interest in the presence of long-time "tails" in time-dependent correlation functions which were once commonly considered to be rapidly decaying. The correlation functions in question are those of the dissipative "fluxes," whose time integrals yield the transport coefficients of linear hydrodynamics, as well as the self-diffusion coefficient. A long-time tail was first observed¹ in a computer study of the singleparticle velocity-autocorrelation function (the flux for self-diffusion) by Alder et al. Following Alder's experiment, there have been numerous theoretical calculations^{2(a)} of the long-time tails. These calculations have usually involved either the mode-mode coupling theory of Kawasaki³ or the Landau-Placzek method, ⁴ and they conclude that, for long times, the flux-autocorrelation functions decay as $t^{-d/2}$, where d is the number of dimensions.

The effect of the t^{-1} tails on the fluxes in two dimensions^{1,2(a)} is to cause logarithmic infinities in the transport coefficients; thus, linearized hydrodynamics "does not exist" in two dimensions. The analogous effect in three dimensions, while less spectacular since the time integrals of the correlation functions remain finite, is still of great importance since the macroscopic equations are nonlocal in time.

Since two-dimensional problems are easier to treat theoretically than three-dimensional problems, we shall confine our attention in this article to the nature of hydrodynamics in two dimensions. We take the attitude that an understanding of the two-dimensional problem is qualitatively similar to an understanding of the three-dimensional problem.

One question with which we shall be concerned is that of the self-consistency of both the LandauPlaczek method, and of the simpler versions of the mode-mode coupling theory, as they have been applied to the long-time tail phenomena. Let us illustrate this point by outlining the mode-mode coupling approach to the long-time tails. Suppose that the density of the one conserved variable in the (hypothetical) system is $A(\mathbf{r}, t)$. We define

$$A_{\vec{\mathbf{k}}}(t) \equiv \int e^{i\vec{\mathbf{k}}\cdot\vec{\mathbf{r}}} A(\vec{\mathbf{r}},t) d\vec{\mathbf{r}}$$
(1)

and

$$\dot{A}_{\vec{k}}(t) \equiv i\vec{k} \cdot \vec{j}_{\vec{k}}(t) , \qquad (2)$$

where j is the flux for A; for small k, the region of interest, the time variation of A becomes very slow $(\dot{A} \rightarrow 0)$. From Mori's⁵ theory of irreversible processes, we may obtain an equation for the correlation function, $\langle A_{\vec{k}}^*(t) A_{-\vec{k}}(0) \rangle$,

$$\int_{0}^{\infty} e^{-st} \langle A_{\vec{k}}(t) A_{-\vec{k}}(0) \rangle dt$$
$$= \frac{\langle A_{\vec{k}} A_{-\vec{k}} \rangle}{s + k^{2} \left[\int_{0}^{\infty} e^{-st} \langle j_{\vec{k}}^{\vec{x}}(t) j_{\vec{k}}^{\vec{x}} \rangle^{\dagger} dt / \langle A_{\vec{k}} A_{-\vec{k}} \rangle \right]}, \quad (3)$$

where $\langle \rangle$ denotes an equilibrium ensemble average,

$$\langle \vec{j}_{\vec{k}}(t)\vec{j}_{-\vec{k}}\rangle^{\dagger} = \langle \left[e^{(1-P_L)i \, \mathcal{L} t} (1-P_L)\vec{j}_{-\vec{k}} \right] (1-P_L)\vec{j}_{-\vec{k}} \rangle$$
$$\equiv \langle \vec{j}_{\vec{k}}(t)\vec{j}_{-\vec{k}}\rangle, \qquad (4)$$

and P_L is the projection operator onto the space of the slow variable $A_{\vec{k}}$,

$$P_{L}G = \left(\left\langle AG \right\rangle / \left\langle A^{2} \right\rangle\right) A \tag{5}$$

for an arbitrary variable G_k^* . In Eq. (5) we have omitted the k subscripts, and we have written $\langle A^2 \rangle$ for $\langle A_k^* A_{-k}^* \rangle$. We shall employ this procedure, from time to time, and we shall omit vector symbols on k subscripts, in cases where our meaning is hopefully obvious. Since A is the only slow variable in the system, and since all the quantities in the correlation function in the denominator of Eq. (3) are projected orthogonal to A, it is usually

1384

assumed⁵ that $\langle \tilde{\mathbf{j}}_{\vec{k}}(t) \, \tilde{\mathbf{j}}_{-\vec{k}} \rangle^{\dagger}$ decays much more rapidly than $\langle A_k(t) A_{-k} \rangle$ for small k. Under this assumption, one is led to the approximation

$$\int_0^\infty e^{-st} \langle \mathbf{j}_k(t) \mathbf{j}_{-k} \rangle^\dagger dt - \int_0^\infty \langle \mathbf{j}_k(t) \mathbf{j}_{-k} \rangle^\dagger dt , \qquad (6)$$

i.e., the Laplace transform is replaced by its s=0 value. Furthermore, for small k, one generally attempts the substitution

$$\langle \mathbf{j}_{k}(t)\mathbf{j}_{-k}\rangle^{\dagger} - \lim_{k \to 0} \langle \mathbf{j}_{k}(t)\mathbf{j}_{-k}\rangle^{\dagger}$$
 (7)

Under these conditions, we recover the usual $^{\scriptscriptstyle b}$ result

$$\int_{0}^{\infty} e^{-st} \left\langle A_{k}(t) A_{-k} \right\rangle dt = \left\langle A^{2} \right\rangle / (s + k^{2} \lambda)$$
(8a)

$$\langle A_k(t)A_{-k}\rangle = \langle A^2\rangle \, e^{-k^2\lambda t} \,, \tag{8b}$$

where $\boldsymbol{\lambda},$ the transport coefficient, is given by the relation

$$\lambda = \lim_{k \to 0, s \to 0} \int_0^\infty e^{-st} \frac{\langle \tilde{\mathbf{j}}_k^*(t) \, \tilde{\mathbf{j}}_{-k}^* \rangle^{\dagger}}{\langle \mathcal{A}^2 \rangle} \, dt = \lim_{k \to 0, s \to 0} \lambda(k, s).$$
(9)

The validity of Eqs. (8) depends upon the assumption that $\langle \vec{j}_k(t) \vec{j}_{-k} \rangle^{\dagger}$ is rapidly decaying, and the assumption that $\lim_{k \to 0} \langle \vec{j}_k(t) \vec{j}_{-k} \rangle^{\dagger}$ is well behaved.

In the mode-mode coupling theory, ³ one recognizes that, while $\vec{j}_{k}^{\dagger}(t)$ is orthogonal to A_{k} in the sense that $\langle A_{k} \vec{j}_{-k}^{\dagger}(t) \rangle = 0$, we may form a set of bilinear variables, $A_{k+k}, A_{-k'}$, with wave vector \vec{k} for all $\vec{k'}$, which do not have to be orthogonal to $(1 - P_{L}) \vec{j}_{k}$. Since A_{k} is slowly varying for small k, $A_{k+k}, A_{-k'}$, will also be slowly varying if k and k' are sufficiently small. Thus, we may write³

$$(1 - P_L) \vec{j}_k = [(1 - P_L) \vec{j}_k]^{\text{mic}} + \sum_{\vec{k}'}^{\vec{k}_c} \frac{\langle (1 - P) \vec{j}_k A_{-k-k'} A_{k'} \rangle}{\langle (A_{k+k'} A_{-k'})^2 \rangle} A_{k+k'} A_{-k'}, \quad (10)$$

where \vec{k}_c is a "cutoff" wave vector such that A_{k+k}, A_{-k} , is no longer slowly varying for $k' \ge k_c$, and we have assumed that $\langle (A_{k+k}, A_{-k'})(A_{-k-k'}, A_{k'}) \rangle$ is zero unless $\vec{k}' = \vec{k}''$. By slowly varying, we mean slow compared to molecular relaxation times. The quantity $[(1 - P_L) j_k]^{\text{mic}}$ is the microscopic, or rapidly decaying, part of $(1 - P_L) \dot{j}_k$, while we have hopefully isolated the slowly varying part of $(1 - P_L) \vec{j}_k$ in the second term on the right-hand side of Eq. (10). We have ignored trilinear and higher nonlinear parts of the flux, of form $A_{k+k'+k'}, A_{-k'}, A_{-k''}$, etc., and we shall ignore these terms throughout the article; this is our first basic assumption. The consensus of opinion^{1,2(a)} seems to be that the most important effects are contained in the bilinear terms, although this question remains somewhat open.

As a consequence of the above, $\langle j_k(t) j_{-k} \rangle^{\dagger}$ may no longer be considered rapidly decaying, and we must, at the very least, consider the transport coefficient,

 λ [see Eq. (9)], to be a function of frequency. Let us, nonetheless, consider the calculation of $\lim_{s \to 0, k \to 0}$, $\lambda(k, s)$. We have

$$\lambda = \lambda_{\min} + \frac{1}{\langle A^2 \rangle} \lim_{k \to 0} \sum_{k'}^{k_c} \frac{\langle (1 - P_L) \tilde{j}_k^* A_{-k-k'} A_k \rangle^2}{\langle (A_{k+k'}, A_{-k'})^2 \rangle^2} \times \int_0^\infty \langle [A_{k+k'}, A_{-k}](t) [A_{-k-k'}, A_{k'}](0) \rangle^\dagger dt, \quad (11)$$

where λ_{mic} arises from the "microscopic" part of $(1-P_L)\mathbf{j}_k$, and we have ignored any cross correlations between the slowly and rapidly decaying parts of $(1-P_L)\mathbf{j}_k$. If we further assume that the time-independent correlation functions in Eq. (9) are independent of \mathbf{k} and \mathbf{k}' , and if we make the factorization approximation,

$$\begin{split} & \langle \left[A_{k*k}, A_{-k}, \right](t) \left[A_{-(k+k')}A_{k}, \right](0) \rangle \\ & = \langle A_{k*k}, (t)A_{-k-k'} \rangle \langle A_{k'}(t)A_{-k'} \rangle, \end{split}$$

we obtain

$$\lambda = \lambda_{\min} + \frac{1}{\langle A^2 \rangle} \frac{\langle (1 - P_L) j_0^2 A_{k'} A_{-k'} \rangle^2}{\langle \langle A_{k'} A_{-k'} \rangle^2 \rangle^2} \\ \times \int_0^\infty \sum_{k'}^{k_c} \langle A_{k'}(t) A_{-k'} \rangle^{t_2} dt. \quad (12)$$

A comparison of Eqs. (12) and (9) shows that, for long times,

$$\lim_{k \to 0} \langle \mathbf{j}_{k}^{\dagger}(t) \mathbf{j}_{-k}^{\dagger} \rangle^{\dagger} \alpha \sum_{\vec{k}'}^{k_{c}} \langle A_{k'}(t) A_{-k'} \rangle^{\dagger 2}.$$
(13)

If we substitute Eq. (8b) into the above, ignoring the \dagger on the correlation function, and replace $\sum_{k=0}^{k} by \left[V/(2\pi)^d \right] \int_0^{k} d\vec{k}'$, where V is the volume and dthe dimensionality, we obtain, for d=2, the muchdiscussed^{1,2(a)} result

$$\lim_{k \to 0} \langle \vec{j}_{k}(t) \vec{j}_{-k} \rangle^{\dagger} \alpha t^{-1}$$
(14)

for long times. As a consequence of Eq. (14), λ , as defined in Eq. (9), is logarithmically infinite in two dimensions. These results depend upon the use of Eq. (8b), a small-k result, for $\langle A_k, (t)A_{-k'} \rangle$ over the whole range of $k' \leq k_c$.

The self-consistency question^{1,2(a)} mentioned earlier is now visible; Eq. (14), which predicts an infinite value for λ , was derived using Eq. (8b), which assumes a finite value for λ . The laws governing the behavior of the slowly varying variable A_k are not at all obvious, in light of the infinite λ predicted by Eq. (14). Since the infinite λ which we have calculated is actually the $\lim_{k,s\to 0}$ of the more general k and s dependent quantity defined in Eq. (9), one is led to the conclusion that $\lambda(k,s)$ is not well behaved for small k and s. Thus, Eqs. (8) which depend both upon the assumption that λ is not a function of s at small s, and the as-

or

sumption that we may take the $\lim_{k\to 0}$, are upon shaky foundations indeed. The calculations outlined above are not sufficiently precise to determine the validity of ordinary linear hydrodynamics, the validity of the *k* expansion, the presence of the $t^{-d/2}$ tails, or the existence of infinite transport coefficients in two dimensions.

Recently, Zwanzig^{2(b)} used the phenomenological, bilinear Navier-Stokes equation to show that the flux correlation function for viscosity has a t^{-1} tail in two dimensions as $k \rightarrow 0$. Zwanzig's method does not suffer from most of the problems outlined above, and his work is similar in spirit to that which we shall present. However, his use of the phenomenological equation, and his early specialization to the $k \rightarrow 0$ limit, indicate that a more complete, nonlinear approach to the problem is desirable.

In this article, we shall deal with all the above considerations via the "complete"³ mode-mode coupling theory. This is done by applying the Mori⁵ formalism with a set of variables consisting of both the linear and bilinear "hydrodynamic" variables. All the fluxes which appear in the resulting theory will therefore be orthogonal to both the linear and the bilinear hydrodynamic variables. As we are ignoring tri- and higher nonlinear hydrodynamic variables, the fluxes will be considered to have no slowly varying parts, and the associated transport coefficients may be considered independent of k and s. We shall then solve the resulting bilinear equations for correlation functions of the linear variables. The results shall allow us to examine the phenomenon displayed in Eq. (14) $(t^{-d/2})$ flux-autocorrelation functions), which has heretofore been derived with so many questionable assumptions.

II. DERIVATION OF EQUATIONS

In this section we consider a classical fluid system consisting of N identical point particles in a volume V. The results are valid in the thermodynamic limit N, $V \rightarrow \infty$, N/V fixed under the assumption that only linear and bilinear combinations of macroscopic variables need be considered.

A. General Theory and Variables

Mori's⁵ identity for the set of variables is

$$\int_{0}^{\infty} e^{-st} \left\langle \underline{\mathbf{Q}}(t) \underline{\mathbf{Q}}^{*} \right\rangle dt = - \underline{\mathcal{T}}^{-1}(s) \int_{0}^{\infty} e^{-st} \left\langle \underline{\mathbf{Q}}(t) \underline{\mathbf{Q}}^{*} \right\rangle dt,$$
(15)

where, e.g., $\langle \dot{\mathbf{Q}}(t)\mathbf{Q}^* \rangle$ is the matrix whose *ij* element is $\langle \dot{\mathbf{Q}}_i(t)\mathbf{Q}^*_j \rangle$, and \mathbf{Q}^*_j is the complex conjugate of \mathbf{Q}_j . The transport matrix $\underline{\gamma}^{-1}(s)$ is given by the relation

$$\underline{\mathcal{T}}^{-1}(s) = \left[-\langle \underline{\dot{\mathbf{Q}}} \mathbf{Q}^* \rangle + \int_0^\infty e^{-st} \langle \underline{\dot{\mathbf{Q}}}(t) \underline{\dot{\mathbf{Q}}}^* \rangle^\dagger dt\right] \langle \underline{\mathbf{Q}} \mathbf{Q}^* \rangle^{-1}.$$
(16)

The significance of the dagger on the time-depen-

dent correlation function on the right-hand side of Eq. (16) has been discussed following Eq. (4); the only modification needed for this many variable case is that the projection operator P projects onto the space of the entire set \underline{Q} . Since the set \underline{Q} is chosen to contain all the linear and bilinear hydrodynamic variables, we shall assume that $\langle \underline{\dot{Q}}(t)\underline{\dot{Q}} \rangle^{\dagger}$ decays on a microscopic time scale. Thus $\underline{T}^{-1}(s)$ may be replaced by its s = 0 value [see discussion concerning Eq. (9)]. We may then take the inverse Laplace transform of Eq. (15) to obtain the equations

$$\frac{d}{dt} \langle \underline{\mathbf{Q}}(t) \underline{\mathbf{Q}}^* \rangle = -\underline{\tau}^{-1} (s=0) \langle \underline{\mathbf{Q}}(t) \underline{\mathbf{Q}}^* \rangle \,. \tag{17}$$

Equations (17) are a set of coupled linear differential equations for the correlation functions. We shall call the coefficients which arise from the first and second terms on the right-hand side of Eq. (16) as the Euler coefficients and the dissipative coefficients, respectively. The long-time tail behavior of $\langle \underline{Q}(t)\underline{Q}^* \rangle$ arises from the fact that, in the thermodynamic limit, \underline{Q} has an infinite number of components and the set of Eqs. (17) consists of an infinite number of equations.

The conserved variables for a simple fluid⁶ are the spatial Fourier transform of the number density,

$$n_k = \sum_{i=1}^{N} e^{i\vec{k}\cdot\vec{r}_i}, \qquad (18a)$$

of the momentum density,

$$\vec{\mathbf{p}}_{k} = \sum_{i=1}^{N} \vec{\mathbf{p}}_{i} e^{i\vec{\mathbf{k}}\cdot\vec{\mathbf{r}}_{i}},\tag{18b}$$

and of the energy density,

$$\epsilon_k = \sum_{i=1}^N \epsilon_i e^{i\vec{k}\cdot\vec{r}_i},\tag{19}$$

where $\vec{\mathbf{r}}_i$, \vec{p}_i , and $\boldsymbol{\epsilon}_i$ are the center-of-mass position, the momentum, and the energy of the *i*th particle, respectively, and N is the number of particles. In the following, for simplicity, we shall ignore $\boldsymbol{\epsilon}_k$; this corresponds roughly to the assumption that there exist no temperature fluctuations in the system, and should not affect the general form of our results. We shall denote the set of linear variables of Eqs. (18) and (19) by $\underline{\mathbf{L}}_k$.

The bilinear variables $\underline{B}_{k,k'}$ are $n_{k+k'}n_{-k'}$, $n_{k+k'}\vec{p}_{-k'}$, $\vec{p}_{k+k'}n_{-k'}$, and $\vec{p}_{k+k'}\vec{p}_{-k'}$, for all $\vec{k}' < \vec{k}_c$. We include both $n_{k+k'}\vec{p}_{-k'}$ and $\vec{p}_{k+k'}n_{-k'}$ as an alternative to keeping just one of the forms, say $n_{k+k_i}\vec{p}_{-k_i}$, with the recognition that both $\vec{k}_i = \vec{k}'$ and $\vec{k}_i = \vec{k} - \vec{k}'$ lead to different variables of form $L_{k+k'}L'_{-k'}$. Of course, a variable $L_{k+k_i}L_{-k_i}$ is identical for $\vec{k}_i = \vec{k}'$ and $\vec{k}_i = -\vec{k} - \vec{k}'$.

The derivation of the bilinear "hydrodynamic" equations is simplified if we use bilinear variables, $\underline{B}'_{k,k'}$ which have been orthogonalized to the linear variables, in the sense that $\langle \underline{B}'_{k,k'}, \underline{L}_{-k} \rangle = 0$. The necessary orthogonalizations may be performed with the aid of the relations⁶

$$\langle n_k n_{-k} \rangle = Ng(k), \tag{20}$$

where g(k) is of order unity and involves a pair distribution function, and

$$\langle \vec{\mathbf{p}}_i \vec{\mathbf{p}}_i \rangle = m k_B T \,\delta_{ij} \,\mathbf{I} \,, \tag{21}$$

where <u>I</u> is the unit tensor, δ_{ij} is the Kroneker δ , T is the absolute temperature, k_B is Boltzmann's constant, and m is the particle mass. We also require the definition

$$\langle n_{k+k}, n_{-k}, n_{-k} \rangle = Nf(k, k'), \qquad (22)$$

where f(k, k') is of the order of unity, and involves a three-particle distribution function. The orthogonalizations are

$$(\vec{\mathbf{p}}_{k+k}, \vec{\mathbf{p}}_{-k})' = (\vec{\mathbf{p}}_{k+k}, \vec{\mathbf{p}}_{-k}) - mk_B T n_k \underline{\mathbf{I}}, \qquad (23a)$$

$$(n_{k+k}, \vec{p}_{-k'})' = (n_{k+k}, \vec{p}_{-k'}) - g(k+k')\vec{p}_{k}, \qquad (23b)$$

$$(\vec{p}_{k+k}, n_{-k'})' = (\vec{p}_{k+k}, n_{-k'}) - g(k')\vec{p}_k, \qquad (23c)$$

$$(n_{k+k}, n_{-k'})' = (n_{k+k}, n_{-k'}) - [f(k, k')/g(k)]n_k.$$
 (23d)

The $\underline{B'}$ variables, plus the linear variables, constitute our "complete set" of slowly varying variables. Hereafter, we shall refer only to the primed bilinear variables, and we shall therefore omit the primes.

B. Euler Equations: Schematic

We now use Eq. (16) to derive the bilinear Euler equations. First we shall give a general discussion of the order of magnitude in N, the number of particles, of the elements of the matrices $\langle \dot{\mathbf{Q}} \mathbf{Q}^* \rangle$, $\langle \underline{\mathbf{Q}} \underline{\mathbf{Q}}^* \rangle$, $\langle \underline{\mathbf{Q}} \underline{\mathbf{Q}}^* \rangle^{-1}$, and the Euler $\underline{\mathcal{T}}^{-1}$ matrix, $\overline{\langle \underline{\mathbf{Q}} \underline{\mathbf{Q}}^* \rangle}$ $\langle \overline{Q} \, \overline{Q}^* \rangle^{-1}$; \overline{N} becomes infinite in the thermodynamic limit, and is an important ordering parameter. We wish to determine which coefficients may be discarded, as well as to outline the detailed derivation which is to follow. The static average of a product of two linear variables, or of the product of the time derivative of a linear variable with a linear variable, may be of order N; these same averages, for bilinear variables with the same \vec{k}' , may be of order N^2 . The static average of the product of a bilinear variable with a linear variable, a bilinear variable with a different \mathbf{k}' , or the derivative of a bilinear variable with a different \vec{k}' , may be of order N. We shall arrange the components of the vector Q with the linear variables first, followed by the bilinear variables for a given $\vec{k'}$, followed by the bilinear variables for a different $\vec{k'}$, and so on. Then, the $\langle \dot{Q} Q^* \rangle$ and $\langle Q Q^* \rangle$ matrices have a diagonal block in the upper left-hand corner which may be of order N, a further set of

diagonal blocks (for each \vec{k}') which may be of order N^2 , and off-diagonal (block) elements which may be of order N. Owing to our orthogonalization, there are no linear-bilinear coefficients in $\langle Q Q^* \rangle^{-1}$. It may then be seen from an expansion in 1/N that $\langle Q Q^* \rangle^{-1}$ has diagonal blocks of order 1/N and $1/N^2$, with off-diagonal elements of order $1/N^3$.

We may now schematically carry out the multiplication, $\langle \underline{\hat{Q}} \mathbf{Q}^* \rangle \langle \underline{\mathbf{Q}} \mathbf{Q}^* \rangle^{-1}$, to obtain the Euler equations. It is necessary to realize that, while $N \rightarrow \infty$ in the thermodynamic limit, so does M, the number of values of $\mathbf{\hat{k}}'$ between 0 and $\mathbf{\hat{k}}_c$, the cutoff wave vector. Thus, some of the elements of the product matrix, $\langle \underline{\mathbf{Q}} \mathbf{Q}^* \rangle \langle \underline{\mathbf{Q}} \mathbf{Q}^* \rangle^{-1}$, will consist of Mterms of a given order in N, and the total N order of the matrix element will be M times the N order of the individual terms. The number M is given by

$$M = \sum_{k'=0}^{k_c} 1 \approx V k_c^d, \qquad (24)$$

where we have used the substitution

$$\Sigma \to \frac{V}{(2\pi)^d} \int d^d \vec{\mathbf{k}},$$

V is the volume, and d is the number of dimensions If we let $V = Na^{d}$, where a is a length on the order of the interparticle spacing,

$$M \approx N(ak_c)^d. \tag{25}$$

It is a basic requirement of our approach that \bar{k}_c is a wave vector with the property that $(ak_c)^d \ll 1$. With this fact in mind, we obtain the "schematic" Euler equations

$$\frac{d}{dt} \langle \underline{\mathbf{L}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle = 1 \langle \underline{\mathbf{L}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle + \frac{1}{N} \sum_{\mathbf{k}1} \langle \underline{\mathbf{B}}_{k,k} \mathbf{1}(t) \underline{\mathbf{Q}}_{-k} \rangle,$$
(26a)
$$\frac{d}{dt} \langle \underline{\mathbf{B}}_{k,k} \mathbf{1}(t) \underline{\mathbf{Q}}_{-k} \rangle = 1 \langle \underline{\mathbf{L}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle + 1 \langle \underline{\mathbf{B}}_{k,k} \mathbf{1}(t) \underline{\mathbf{Q}}_{-k} \rangle + \frac{1}{N} \sum_{\mathbf{k}^{2} + \mathbf{k}^{2} \mathbf{1}} \langle \underline{\mathbf{B}}_{k,k} \mathbf{2}(t) \underline{\mathbf{Q}}_{-k} \rangle,$$
(26b)

where <u>L</u> and <u>B</u> represent linear and bilinear variables, respectively, and Q_{-k} may be any member of the set of variables. Equations (26) are merely intended to indicate the N order of the coefficients in the coupled equations; although Eqs. (26) were derived with the Euler equations in mind, they are in fact correct for the complete (Euler plus dissipative) equations.

Consider the solution, via Laplace transform, of the schematic equations

$$\langle \underline{\mathbf{L}}_{k}(s) \, \underline{\mathbf{Q}}_{-k} \rangle \, \langle s+1 \rangle = \langle \underline{\mathbf{L}}_{k} \underline{\mathbf{Q}}_{-k} \rangle + \frac{1}{N} \sum_{\underline{g}^{1}} \langle \underline{\mathbf{B}}_{k,k} \mathbf{1}(s) \, \underline{\mathbf{Q}}_{-k} \rangle \,,$$

$$\langle \underline{\mathbf{B}}_{k,k} \mathbf{1}(s) \, \underline{\mathbf{Q}}_{-k} \rangle \langle s+1 \rangle = \langle \underline{\mathbf{B}}_{k,k} \mathbf{1} \, \underline{\mathbf{Q}}_{-k} \rangle + \langle \underline{\mathbf{L}}_{k}(s) \, \underline{\mathbf{Q}}_{-k} \rangle$$

$$\langle \underline{\mathbf{P}}_{k,k} \mathbf{1}(s) \, \underline{\mathbf{Q}}_{-k} \rangle \langle s+1 \rangle = \langle \underline{\mathbf{P}}_{k,k} \mathbf{1} \, \underline{\mathbf{Q}}_{-k} \rangle + \langle \underline{\mathbf{L}}_{k}(s) \, \underline{\mathbf{Q}}_{-k} \rangle$$

$$-\frac{1}{N}\sum_{\substack{k^{2}\neq k^{1}}} \langle \mathbf{B}_{k,k^{2}}(s) \mathbf{Q}_{-k} \rangle, \quad (27b)$$

where $\langle \underline{\mathbf{Q}}(s) \underline{\mathbf{Q}}^* \rangle$ is the Laplace transform of $\langle \underline{\mathbf{Q}}(t) \underline{\mathbf{Q}}^* \rangle$ As we wish to obtain the correlation functions of the linear variables, we let $\underline{\mathbf{Q}}_{-k}$ in Eqs. (27) be $\underline{\mathbf{L}}_{-k}$. Combining Eqs. (27), and using the orthogonality of $\underline{\mathbf{L}}$ and $\underline{\mathbf{B}}'$, we obtain

$$\langle \underline{\mathbf{L}}_{k}(s) \underline{\mathbf{L}}_{-k} \rangle \left(s + 1 + \frac{1}{N} \sum_{\mathbf{k}^{1}} \frac{1}{s+1} \right) = \langle \underline{\mathbf{L}}_{k} \underline{\mathbf{L}}_{-k} \rangle$$

$$+ \frac{1}{N^{2}} \sum_{\mathbf{k}^{1}} \sum_{\mathbf{k}^{2} \neq \mathbf{k}^{1}} \frac{1}{s+1} \langle \underline{\mathbf{B}}_{k,k}^{2}(s) \underline{\mathbf{L}}_{-k} \rangle.$$
(28)

The $1/N \sum_{k=1}^{r} [1/(s+1)]$ term, which multiplies $\langle \underline{L}_{k}(s) \underline{L}_{-k} \rangle$ on the left-hand side of Eq. (28), represents a contribution to the frequency spectrum of $\langle \underline{L}_{k}(s) \underline{L}_{-k} \rangle$ due to linear-nonlinear "coupling," which is of order $(ak_{c})^{d}$, and which therefore remains finite as $N \to \infty$. To obtain the first contribution to the spectrum of $\langle \underline{L}_{k}(s) \underline{L}_{-k} \rangle$ due to <u>coupling</u> among nonlinear variables of different k', we combine Eqs. (28) and (27b), with the result

$$\langle \underline{\mathbf{L}}_{k}(s) \underline{\mathbf{L}}_{-k} \rangle \left(s + 1 + \frac{1}{N} \sum_{\vec{k}^{1}} \frac{1}{s+1} + \frac{1}{N^{2}} \sum_{\vec{k}^{1}} \sum_{\vec{k}^{2} \neq \vec{k}^{1}} \frac{1}{s+1} \right)$$

$$= \langle \underline{\mathbf{L}}_{k} \underline{\mathbf{L}}_{-k} \rangle + \frac{\langle a k_{c} \rangle^{d}}{N^{3}} \sum_{\vec{k}^{1}} \sum_{\vec{k}^{2} \neq \vec{k}^{1}} \sum_{\vec{k}^{3} \neq \vec{k}^{2}, \vec{k}^{1}} \frac{\langle \underline{\mathbf{B}}_{k,k}^{3}(s) \underline{\mathbf{L}}_{-k} \rangle}{(s+1)^{2}} .$$

$$(29)$$

By repeating the above process, we see that the contribution to the spectrum of $\langle \underline{L}_k(s)\underline{L}_{-k}\rangle$ due to coupling among bilinear variables of different \overline{k}' is of order $(ak_c)^{2d}[1+(ak_c)^{2d}+(ak_c)^{4d}\cdots]$, i.e., of order $(ak_c)^{2d}\sum_{n=0}^{\infty}[(ak_c)^{2d}]^n$. The sum over *n* is just a geometric series, and thus the coupling among nonlinear variables of different \overline{k}' contributes a part of order $(ak_c)^{2d}$ to the spectrum of $\langle \underline{L}_k(s)\underline{L}_{-k}\rangle$, while the contribution of linear-nonlinear coupling is of order $(ak_c)^d$. The $\underline{B}_{k'} - \underline{B}_{k''}$ coupling is therefore a "higher order" effect which we shall neglect in all the following. Since the only effect of the off-diagonal elements of the inverse matrix, $\langle \underline{Q} \underline{Q}^* \rangle^{-1}$, is to produce the $\underline{B}_{k'} - \underline{B}_{k''}$ coupling, we may then assume that $\langle \underline{Q} \underline{Q}^* \rangle^{-1}$ is (block) diagonal.

C. Euler Equations

It is now a simple matter to calculate $\langle \underline{Q} \underline{Q}^* \rangle$, and, by inversion of each diagonal block, $\langle \underline{Q} \underline{Q}^* \rangle^{-1}$. We need the relation⁶

$$\langle n_k \vec{\mathbf{p}}_{-k} \rangle = 0,$$
 (30)

as well as the properties of averages of various products of single-particle momenta, which follow from the Maxwellian distribution. We also require the factorization approximation for equal time correlation functions, which is correct to order 1/N (away from the critical point), and sufficient for our purposes:

$$\langle \langle A_{k+k}, B_{-k}, \rangle \rangle \langle C_{-k-k}, D_{k}, \rangle \rangle = \langle A_{k+k}, C_{-k-k}, \rangle \langle D_{k}, B_{-k}, \rangle$$

$$+ \langle A_{k+k}, D_{k}, \rangle \langle B_{-k}, C_{-k-k}, \rangle.$$
(31)

The second term on the right-hand side of Eq. (31) contributes only if $\vec{k}' = -\frac{1}{2}\vec{k}$.

We shall assume that g(k) or g(k') may always be replaced by $g(k \rightarrow 0)$, which we shall write as g. The argument of g(k) is always less than k_c , and, away from the critical point, g(k) does not differ significantly from $g(k \rightarrow 0)$ until k^{-1} is of the order of the range of the intermolecular potential. Similar considerations hold for f(k, k') [see Eq. (22)]. With the above information, it is seen that $\langle QQ^* \rangle$ is not only block diagonal; it may, with negligible error (the error is of order 1/N, with no compensating factors of M), be considered diagonal. The Euler equations may thus be written

$$\frac{d}{dt} \langle Q_i(t) Q_j^* \rangle = \sum_{l} \frac{\langle Q_l Q_l^* \rangle}{\langle Q_l Q_l^* \rangle} \langle \dot{Q}_i(t) Q_j^* \rangle .$$
(32)

All the elements of $\langle \dot{\mathbf{Q}} \mathbf{Q}^* \rangle$ follow easily from the information presented above, the conservation laws, and the relation

$$\langle \dot{A}B \rangle = - \langle A\dot{B} \rangle.$$
 (33)

The first of the resulting equations is

$$\frac{d}{dt} \langle n_{k}(t)\underline{\mathbf{Q}}_{-k} \rangle = \frac{i\overline{\mathbf{k}}}{m} \cdot \langle \vec{\mathbf{p}}_{k}(t)\underline{\mathbf{Q}}_{-k} \rangle, \qquad (34)$$

which is the equation of continuity. The bilinear Euler momentum transport equation is

$$\frac{d}{dt} \langle \vec{\mathbf{p}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle = i\vec{\mathbf{k}} \frac{k_{B}T}{g} \langle u_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle + i\vec{\mathbf{k}} \frac{k_{B}T}{2Ng} \left(1 - \frac{f}{g^{2}} \right)$$

$$\times \sum_{\vec{\mathbf{k}}'} \langle [n_{k+k} \cdot n_{-k'}](t) \underline{\mathbf{Q}}_{-k} \rangle + \frac{1}{2Nm} \sum_{\vec{\mathbf{k}}'} \langle [i\vec{\mathbf{k}} \cdot \vec{\mathbf{p}}_{k+k'} \cdot \vec{\mathbf{p}}_{-k'} + \vec{\mathbf{p}}_{k+k'} \cdot \vec{\mathbf{p}}_{-k'} \cdot i\vec{\mathbf{k}}](t) \underline{\mathbf{Q}}_{-k} \rangle. \tag{35}$$

The factor $\frac{1}{2}$ preceding the $\sum_{\mathbf{k}}$'s prevents overcounting via the symmetry of \mathbf{k}' and $-\mathbf{k} - \mathbf{k}'$.

In order to discuss Eq. (35), recall that, near equilibrium, the equations governing the dynamics of correlation functions are believed⁷ to be identical to those governing the dynamics of nonequilibrium ensemble averages of the slow variables. The heuristic, nonlinear Navier-Stokes equation is⁸

$$\frac{d}{dt} \langle \vec{p}(\vec{\mathbf{r}},t) \rangle' = \vec{\nabla} \cdot \underline{\boldsymbol{\varphi}} (\vec{\mathbf{r}},t) + \vec{\nabla} \cdot \langle \vec{p}(\vec{\mathbf{r}},t) \vec{\nabla} (\vec{\mathbf{r}},t) \rangle', \quad (36)$$

where $\langle \rangle'$ denotes a nonequilibrium ensemble average, σ is the pressure tensor, and \vec{v} is the velocity field. We may inverse Fourier transform Eq. (35) to obtain

$$\frac{d}{dt} \left\langle \vec{p}(\vec{\mathbf{r}},t) \underline{\mathbf{Q}}(\mathbf{0},\mathbf{0}) \right\rangle = \frac{k_B T}{g} \vec{\nabla} \left\langle \delta n(\vec{\mathbf{r}},t) \underline{\mathbf{Q}} \right\rangle$$

$$+\frac{k_{B}T}{g}\left(1-f/g^{2}\right)\vec{\nabla}\left\langle\left[\delta n^{2}(\vec{\mathbf{r}},t)/n_{0}\right]\underline{Q}\right\rangle$$
$$+\vec{\nabla}\cdot\left\langle\left[\vec{p}(\vec{\mathbf{r}},t)\vec{p}(\vec{\mathbf{r}},t)/n_{0}m\right]\underline{Q}\right\rangle,\quad(37)$$

where n_{θ} is the average number density and δn is the density fluctuation. This result is correct in a coarse-grained sense, i.e., for lengths $\geq k_c^{-1}$. This limitation should not be a factor in comparing Eqs. (36) and (37), as the heuristic, macroscopic Eq. (36) is itself only intended for use over macroscopic distance scales. One compares Eqs. (36) and (37) by realizing that a term $\langle Q(\vec{r}, t) Q \rangle$ in Eq. (37) corresponds to a term $\langle Q(\vec{r}, t) \rangle'$ in Eq. (36). It is well known that the first term on the righthand side of Eq. (37) corresponds to the linear term in the expansion in the density fluctuations of the pressure gradient, which appears on the right-hand side of Eq. (36), i.e.,

$$\frac{k_{p}T}{g}\vec{\nabla}n(\vec{\mathbf{r}},t) = \vec{\nabla}\cdot \frac{d\,\varrho_{0}}{dn_{0}} \quad n(\vec{\mathbf{r}},t).$$
(38)

The second term on the right-hand side of Eq. (37) is easily shown to correspond to the quadratic term in the expansion of the pressure in the density. The velocity field $\vec{v}(\vec{r})$ is related to our variables by the relation⁸

$$\vec{\mathbf{v}}(\vec{\mathbf{f}}) = \vec{\mathbf{p}}(\vec{\mathbf{f}}) / mn(\vec{\mathbf{f}}). \tag{39}$$

If we write

$$n(\vec{\mathbf{r}}) = n_{\bullet} + \delta n(\vec{\mathbf{r}}), \tag{40}$$

and if we combine Eqs. (40), (39), and (36), and expand in powers of the density fluctuations, we obtain

$$\frac{d}{dt} \langle \boldsymbol{p}(\vec{\mathbf{r}}, t) \rangle' = \vec{\nabla} \cdot \underline{\boldsymbol{\sigma}}(\vec{\mathbf{r}}, t) + \vec{\nabla} \cdot \left(\frac{\langle \vec{\mathbf{p}}(\vec{\mathbf{r}}, t) \vec{\mathbf{p}}(\vec{\mathbf{r}}, t) \rangle'}{n_0 m} - \frac{\langle \vec{\mathbf{p}}(\vec{\mathbf{r}}, t) \vec{\mathbf{p}}(\vec{\mathbf{r}}, t) \delta n \rangle'}{n_0^2 m} + \cdots \right). \quad (41)$$

The first term in the large parentheses on the righthand side of Eq. (41) corresponds to the bilinear momentum term which we have derived in Eq. (37). Thus, we see that simple expansions of the heuristic Eq. (36), to bilinear order in the fluctuations, generate the Euler equation which we have derived independently. One should recall that the bilinear variables which appear in Eq. (37) are actually the orthogonalized variables defined in Eqs. (23).

There exist no heuristic analogues to our Euler equations for the bilinear variables. These equations are

$$\frac{d}{dt} \langle [n_{k+k}, n_{-k'}](t) \underline{\mathbf{Q}}_{-k} \rangle = \frac{(g - f/g)}{m} i \overline{\mathbf{k}} \cdot \langle \overline{\mathbf{p}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle + \frac{i (\overline{\mathbf{k}} + \overline{\mathbf{k}}')}{m} \cdot \langle [\overline{\mathbf{p}}_{k+k'}, n_{-k'}](t) \underline{\mathbf{Q}}_{-k} \rangle$$

$$-\langle [n_{\vec{k}+\vec{k}}, \vec{p}_{-\vec{k}}, (i\vec{k}'/m)](t)Q_{-\vec{k}}\rangle, (42)$$

$$\frac{d}{dt} \langle [n_{k+k}, \mathbf{\tilde{p}}_{-k}](t)\mathbf{Q}_{-k} \rangle = -\frac{k_B T}{g} i\mathbf{\tilde{k}}' \langle [n_{\mathbf{\tilde{k}}+\mathbf{\tilde{k}}'}, n_{-\mathbf{\tilde{k}}'}](t)\mathbf{Q}_{-k} \rangle \\ + \frac{i(\mathbf{\tilde{k}}+\mathbf{\tilde{k}}')}{m} \cdot \langle [\mathbf{\tilde{p}}_{k+k}, \mathbf{\tilde{p}}_{-k}](t)\mathbf{Q}_{-k} \rangle, \quad (43)$$
$$\frac{d}{dt} \langle [\mathbf{\tilde{p}}_{k+k}, \mathbf{\tilde{p}}_{-k}](t)\mathbf{Q}_{-k} \rangle = k_B T \langle [i\mathbf{\tilde{k}}\mathbf{\tilde{p}}_{k}(t) + \mathbf{\tilde{p}}_{k}(t)i\mathbf{\tilde{k}}]\mathbf{Q}_{-k} \rangle \\ + (k_B T/g)i(\mathbf{\tilde{k}}+\mathbf{\tilde{k}}') \langle [n_{k+k}, \mathbf{\tilde{p}}_{-k}](t)\mathbf{Q}_{-k} \rangle \\ - (k_B T/g) \langle [\mathbf{\tilde{p}}_{k+k}, n_{-k}, i\mathbf{\tilde{k}}'](t)\mathbf{Q}_{-k} \rangle. \quad (44)$$

The equation for $\langle [\vec{p}_{k+k}, n_{-k'}](t) \mathbf{Q}_{-k} \rangle$ follows from Eq. (43) via the substitution $\vec{k'} \rightarrow -\vec{k} - \vec{k'}$.

D. Dissipative Coefficients

We now treat the dissipative coefficients, i.e., the coefficients arising from the second term on the right-hand side of Eq. (16). Our main aim here is to demonstrate the origin of the $t^{-d/2}$ tails of the correlation functions. Therefore, while we shall be careful in treating the physics of the problem, we shall be somewhat cavalier in handling the algebra involved.

Our basic physical assumption is that the set \underline{Q} , which consists of linear and bilinear forms, contains all of the pertinent slow variables in the system. Thus, while a k expansion of the dissipative terms in which only linear forms are included is not valid, a k and k' expansion for the set \underline{Q} is valid in two and three dimensions. We shall retain terms up to quadratic order (k^2, kk', k^2) in these variables.

It is necessary to obtain the correlation functions $\langle (1-P)\dot{\mathbf{Q}}e^{(1-P)i\mathfrak{L}t}(1-P)\dot{\mathbf{Q}}\rangle$ [see Eq. (4)]. Since $\mathbf{\tilde{p}}_k$ and $n_{k*k'}\mathbf{\tilde{p}}_{-k'}$ are contained in our set of variables, and since $\dot{n}_k = i\mathbf{k}_m \cdot \mathbf{\tilde{p}}_k$ and

$$\frac{d}{dt} n_{k+k'} n_{-k'} = \frac{i(\vec{k} + \vec{k}')}{m} \cdot \vec{p}_{k+k'} n_{-k'} - \frac{i\vec{k}'}{m} \cdot n_{k+k'} \vec{p}_{-k'},$$

it is easy to see that $(1 - P)n_k = 0$ and $(1 - P) \times (n_{k+k}, n_{-k},)' = 0$. Furthermore,

$$(1-P)\frac{d}{dt}\left(n_{k+k}, \mathbf{p}_{-k'}\right) = (1-P)(i\mathbf{k}' \cdot n_{k+k}, \mathbf{\sigma}_{-k'}), \quad (45)$$

where the stress tensor $\sigma_{\mathbf{k}}$ is defined by the relation

$$\dot{\vec{\mathbf{p}}}_{k} = i\vec{\mathbf{k}}\cdot\underline{\sigma}_{k}.$$
(46)

There are no obvious subtractions, similar to those described above, owing to the action of (1-P) upon $(d/dt)\vec{p}_k$ or $(d/dt)(\vec{p}_{k+k'}\vec{p}_{-k'})$. Of course, (1-P) is expected to remove any linear or bilinear hydrodynamic contributions to \dot{Q} , allowing us to assume that the dissipative coefficients are independent of frequency and expandable in k and k'.

First consider the \vec{p}_k , \vec{p}_k dissipative coefficient. We have

$$\lim_{s\to 0}\int_0^\infty e^{-st}\langle \dot{\vec{p}}_k(t)\dot{\vec{p}}_{-k}\rangle^\dagger dt = \int_0^\infty dt (-i\vec{k})\cdot\langle (1-P)\underline{\sigma}_{-k}e^{(1-P)i\mathcal{L}t}(1-P)\underline{\sigma}_{+k}\rangle \cdot (i\vec{k}) = Nmk_BT \cdot i\vec{k}\cdot\underline{\alpha}^{(4)}\cdot -i\vec{k}, \quad (47)$$

where $\underline{\alpha}^{(4)}$ is the "bare" kinematic viscosity tetrad; $\underline{\alpha}^{(4)}$ differs from the ordinary kinematic viscosity tetrad insofar as it has no bilinear hydrodynamic contributions. The tetrad $\underline{\alpha}^{(4)}$ is related to the bare coefficients of kinematic shear and bulk viscosity, η^0 and η^0_B , respectively, by the relation

$$(i\vec{k}\cdot\underline{\alpha}^{(4)}\cdot -i\vec{k})\cdot\vec{p}_{k}=i\vec{k}\cdot\left\{\eta^{0}[i\vec{k}\cdot\vec{p}_{k}+\vec{p}_{k}i\vec{k}-\frac{2}{3}i\vec{k}\cdot\vec{p}_{k}\vec{1}]\right.$$
$$\left.+\eta^{0}_{B}i\vec{k}\cdot\vec{p}_{k}\vec{1}\right\}, \quad (48)$$

where \vec{I} is the unit tensor. In a similar fashion, we define

$$\lim_{s \to 0} \int_{0}^{\infty} e^{-st} \left\langle (1-P) \frac{d}{dt} \left(n_{-k-k} \cdot \vec{\mathbf{p}}_{+k} \cdot \right)' e^{(1-P) i \mathfrak{L} t} (1-P) \frac{d}{dt} \left(n_{k+k} \cdot \vec{\mathbf{p}}_{-k'} \right)' \right\rangle dt = \int_{0}^{\infty} dt \, i \vec{\mathbf{k}} \cdot \left\langle (1-P) (n_{-k-k} \cdot \underline{\sigma}_{-k'}) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_{-k-k} \cdot \underline{\sigma}_{-k'} \right) e^{(1-P) i \mathfrak{L} t} (1-P) \left(n_$$

It is not necessary to consider the orthogonalized form of the <u>B</u> variables in Eq. (49), and in similar equations, as the effects of the orthogonalizations are of order 1/N. The decay of the correlation function appearing in Eq. (49) might be expected to resemble the decay of the σ autocorrelation function, as n_k is slowly varying. Thus, $\underline{\alpha}^{(4)}$ and $\underline{\beta}^{(4)}$ might be simply related; this point requires further study. Finally, we define the remaining diagonal matrix element,

$$\lim_{s \to 0} \int_{0}^{\infty} e^{-st} \left\langle (1-P) \frac{d}{dt} (\vec{p}_{-k-k}, \vec{p}_{+k},)' e^{(1-P)i\hat{k}t} (1-P) \frac{d}{dt} (\vec{p}_{k+k}, \vec{p}_{-k'}) \right\rangle dt = (Nmk_{B}T)^{2} [i(\vec{k} + \vec{k}') \cdot \underline{\gamma}^{(6)} \cdot - i(\vec{k} + \vec{k}') - i(\vec{k} + \vec{k}') \cdot \underline{\gamma}^{(6)} \cdot i\vec{k}' + i(\vec{k} + \vec{k}') \cdot \underline{\epsilon}^{(6)} \cdot i\vec{k}' + i\vec{k}' \cdot \underline{\epsilon}^{(6)} \cdot i(\vec{k} + \vec{k}')].$$
(50)

It is expected that the sixth-order tensor $\gamma^{(6)}$, like $\underline{\beta}^{(4)}$, should have a close relation to the bare viscosity $\underline{\alpha}^{(4)}$. However, $\underline{\epsilon}^{(6)}$ involves correlation functions equal to zero at t=0, and should be very small.

We shall ignore the off-diagonal dissipative coefficients. Our equations already contain linearbilinear coupling, linear-linear coupling, and bilinear-bilinear coupling which arises from the Euler equations; the coefficients which give rise to this coupling are linear in k and k'. Off-diagonal dissipative coefficients will be bilinear in k and k', and, since we are assuming that k and k' may be treated as small quantities, these coefficients should be neglected. The validity of this procedure shall be explicitly visible later on.

We may now write down the bilinear hydrodynamic equations to second order in k and k':

$$\frac{d}{dt} \langle n_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle = \frac{i\vec{\mathbf{k}}}{m} \cdot \langle \vec{\mathbf{p}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle,$$

$$\frac{d}{dt} \langle \vec{\mathbf{p}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle = i\vec{\mathbf{k}} \frac{k_{B}T}{g} \langle n_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle + i\vec{\mathbf{k}} \frac{k_{B}T}{2Ng} \left(1 - \frac{f}{g^{2}} \right) \sum_{\vec{\mathbf{k}}} \langle \left[n_{k+k} \cdot n_{-k'} \right](t) \underline{\mathbf{Q}}_{-k} \rangle \\
+ \frac{1}{2Nm} \sum_{\vec{\mathbf{k}}} \langle \left[i\vec{\mathbf{k}} \cdot \vec{\mathbf{p}}_{k+k'} \cdot \vec{\mathbf{p}}_{-k'} + \vec{\mathbf{p}}_{k+k'} \cdot \vec{\mathbf{p}}_{-k'} \cdot i\vec{\mathbf{k}} \right](t) \underline{\mathbf{Q}}_{-k} \rangle + (i\vec{\mathbf{k}} \cdot \underline{\alpha}^{(4)} \cdot i\vec{\mathbf{k}}) \cdot \langle \vec{\mathbf{p}}_{k}(t) \underline{\mathbf{Q}}_{-k} \rangle,$$
(51a)

$$\frac{d}{dt}\langle [n_{k+k}, n_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle = \frac{(g - f/g^2)}{m} i\vec{\mathbf{k}} \cdot \langle \vec{\mathbf{p}}_k(t)\underline{\mathbf{Q}}_{-k}\rangle + \frac{i(\vec{\mathbf{k}} + \vec{\mathbf{k}}')}{m} \cdot \langle [\vec{\mathbf{p}}_{k+k}, n_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle - \frac{i\vec{\mathbf{k}}'}{m} \cdot \langle [n_{k+k}, \vec{\mathbf{p}}_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle, \quad (51c)$$

$$\frac{d}{dt}\langle [n_{k+k},\vec{\mathbf{p}}_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle = -\frac{k_BT}{g}i\vec{\mathbf{k}}'\langle [n_{k+k},n_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle + i\frac{(\vec{\mathbf{k}}+\vec{\mathbf{k}}')}{m}\cdot\langle [\vec{\mathbf{p}}_{k+k},\vec{\mathbf{p}}_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle + (i\vec{\mathbf{k}}'\cdot\underline{\boldsymbol{\beta}}^{(4)}\cdot i\vec{\mathbf{k}}')\cdot\langle [n_{k+k},\vec{\mathbf{p}}_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle$$
(51d)

$$\frac{d}{dt} \langle \left[\vec{p}_{k+k}, \vec{p}_{-k} \right] (t) \underline{Q}_{-k} \rangle = k_B T \left[i\vec{k} \langle \vec{p}_k(t) \underline{Q}_{-k} \rangle + \langle \vec{p}_k(t) (i\vec{k}) \underline{Q}_{-k} \rangle \right]$$

$$+\frac{k_{B}T}{g}\left[i(\vec{\mathbf{k}}+\vec{\mathbf{k}}')\langle[n_{k*k'}\vec{\mathbf{p}}_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle-\langle[\vec{\mathbf{p}}_{k*k'}n_{-k'}(i\vec{\mathbf{k}}')](t)\underline{\mathbf{Q}}_{-k}\rangle\right]$$

+
$$\left[i(\vec{\mathbf{k}}+\vec{\mathbf{k}}')\cdot\underline{\gamma}^{(6)}\cdot i(\vec{\mathbf{k}}+\vec{\mathbf{k}}')+i\vec{\mathbf{k}}'\cdot\underline{\gamma}^{(6)}\cdot i\vec{\mathbf{k}}'-i(\vec{\mathbf{k}}+\vec{\mathbf{k}}')\cdot\underline{\epsilon}^{(6)}\cdot i\vec{\mathbf{k}}'-i\vec{\mathbf{k}}'\cdot\underline{\epsilon}^{(6)}\cdot i(\vec{\mathbf{k}}+\vec{\mathbf{k}}')]\cdot\langle[\vec{\mathbf{p}}_{k*k'}\vec{\mathbf{p}}_{-k'}](t)\underline{\mathbf{Q}}_{-k}\rangle.$$
(51e)

Equations (51) are based upon our two fundamental assumptions; the assumption that trilinear and higher-linear variables may be ignored; and the assumption that k and k' may be treated as small. The solution of Eqs. (51) for the autocorrelation functions of the linear variables will constitute, within the context of our assumptions, a self-consistent treatment of the problems discussed in the introduction. We remark that, although the remainder of our work shall be concerned with two dimensions, Eqs. (51) are also valid in three dimensions.

III. SOLUTIONS

It is not possible to answer the questions which interest us by just examining Eqs. (51); the equations must be solved. We shall discuss the properties of the transverse momentum autocorrelation function in two dimensions from Eqs. (51). This discussion illustrates all the general features of nonlinear effects upon hydrodynamic correlation functions in two dimensions. Thus, we let $\vec{k} = k\vec{x}$, and we calculate $\langle p_k^{\nu}(t)p_{k}^{\nu}\rangle$. As we shall proceed by Laplace transform methods, the quantity which we shall actually obtain is

$$\langle p_{k}^{y}(s)p_{-k}^{y}\rangle = \int_{0}^{\infty} e^{-st} \langle p_{k}^{y}(t)p_{-k}^{y}\rangle dt.$$
 (52)

For comparison's sake, we first mention the predictions⁸ of ordinary linear hydrodynamics for $\langle p_{k}^{*}(t)p_{k}^{*}\rangle$; they are $(\vec{k} = k\hat{\vec{x}})$

$$\langle p_{k}^{y}(t)p_{-k}^{y}\rangle\langle p_{k}^{y}p_{-k}^{y}\rangle^{-1} = e^{-k^{2}\eta t}, \qquad (53a)$$

$$\int_0^\infty e^{-st} \langle p_k^{\mathbf{y}}(t) p_{-k}^{\mathbf{y}} \rangle \langle p_k^{\mathbf{y}} p_{-k}^{\mathbf{y}} \rangle^{-1} dt = 1/(s+k^2\eta), \quad (53b)$$

where η is the ordinary coefficient of kinematic shear viscosity. The area under the normalized time-correlation function is equal to $\langle p_k^y(s=0)p_{-k}^y \rangle$ $\times \langle p_k^y p_{-k}^y \rangle^{-1}$, i.e., to $(k^2 \eta)^{-1}$ [see Eq. (53b)].

It is possible to write⁵ an exact equation,

$$\int_{0}^{\infty} e^{-st} \langle p_{k}^{y}(t) p_{-k}^{y} \rangle \langle p_{k}^{y} p_{-k}^{y} \rangle^{-1} dt = 1/[s + k^{2}\eta(k, s)],$$
(54)

where $\eta(k, s)$ is determined as in Eq. (4),

$$\eta(k, s) = \frac{\int_0^\infty e^{-st} \langle (1-P)\dot{p} \frac{y}{2} e^{(1-P)i\hat{L}t} (1-P)\dot{p} \frac{y}{k} \rangle dt}{k^2 \langle p \frac{y}{k} p \frac{y}{2}_k \rangle} .$$
(55)

Equations (53) result from Eqs. (54) and (55) if $\eta(k, s)$ is independent of k and s for small k and s, and $\eta(k, s)$ is replaced by $\lim_{k\to 0, s\to 0} \eta(k, s) = \eta$. After we finish the calculation of $\langle p_k^{y}(s) p_{-k}^{y} \rangle$, we may cast it in the form of Eq. (54) in order to actually determine $\eta(k, s)$; this will allow us to examine the properties of $\lim_{k\to 0,s\to 0} \eta(k, s)$. We may then inverse Laplace transform $\eta(k, s)$ to determine the autocorrelation function of the flux for p_k^y , and to determine if a $t^{-d/2}$ tail exists. Thus, a calculation of $\langle p_k^y(s)p_{-k}^y \rangle$ "includes" a calculation of the hydrodynamic part of the flux autocorrelation function.

Let us now Laplace transform Eq. (51b), for $Q_{-k} = p \frac{\phi}{k}$; the result is

$$\langle p_{k}^{y}(s)p_{-k}^{y}\rangle\langle p_{k}^{y}p_{-k}^{y}\rangle^{-1}$$

$$= \left(s + k^{2}\alpha_{1} - \frac{ik}{Nm}\frac{1}{2}\sum_{k'}\left\{\langle \left[p_{k+k'}^{x}p_{-k'}^{y}\right](s)p_{-k'}^{y}\rangle + \langle \left[p_{k+k'}^{y}p_{-k'}^{z}\right](s)p_{-k'}^{y}\rangle \right\}\langle p_{k}^{y}(s)p_{-k'}^{y}\rangle^{-1}\right)^{-1}, \quad (56)$$

where $\alpha_{\perp} = \eta^0$, the bare shear viscosity discussed earlier. The bilinear contribution to $\eta(k, s)$ [see Eq. (54)] is contained in the third term on the right-hand side of Eq. (56). Our task is to use the linear Eqs. (51) to express the correlation functions involving $p_{k+k}^x p_{-k}^y$ and $p_{k+k}^y p_{-k}^x$ in terms of the p_k^y autocorrelation function, and thus to evaluate the bilinear shear viscosity contribution.

Since Eqs. (51) are rather complicated, we shall employ some simplifying assumptions, and treat some limiting cases, in what is to follow. We emphasize that none of our simplifications will violate any important physical ideas, but will rather be in the nature of neglect of "fine points"; we shall discuss each approximation as it arises. First, we shall calculate $\langle p_{\mu}^{y}(s)p_{-\mu}^{y} \rangle$, and $\eta(k, s)$, for s = 0 and k negligible with respect to k'. Later on we shall see that ignoring k with respect to k'leads to a small error. Next we shall outline the calculations, and present the results, for the two cases, finite k and s = 0, and finite s and $k \ll k'$. In the finite k calculation, we shall assume that the "viscous" tensors, $\alpha^{(4)}$, $\beta^{(4)}$, and $\gamma^{(6)}$ may be replaced by scalars times the unit tensors, $\alpha I^{(4)}$, $\beta I^{(4)}$, and $\gamma I^{(6)}$ and that $\epsilon^{(6)}$ may be equated to zero. These approximations will not affect the essential features of our results. Finally, we shall perform the calculations for finite k and s. under the above mentioned assumptions, and under the assumption that the fluid is incompressible. As we shall see, the assumption of incompressibility leads to a simple multiplicative error in $\eta(k,s)$. owing to overcounting of "shear" modes.

A. Calculation of $\eta(0, 0)$

In order to eliminate correlation functions involving $p_{k+k}^{x} \cdot p_{-k'}^{y}$ and $p_{k+k'}^{y} \cdot p_{-k'}^{x}$ from the right-hand side of Eq. (56), we introduce a very useful transformation. For a given \vec{k}' , consider an (x', y')coordinate frame in which $k' \parallel x'$ (recall $k \parallel x$). We have

$$p^{x}p^{y} + p^{y}p^{x} = 2\cos\phi \sin\phi (p^{x'}p^{x'} - p^{y'}p^{y'}) + (\cos^{2}\phi - \sin^{2}\phi)(p^{x'}p^{y'} + p^{y'}p^{x'}), \quad (57)$$

where ϕ is the angle between \vec{k} and \vec{k}' . We may combine Eqs. (56) and (57), and calculate the correlation functions involving the primed bilinear momenta via the transformed Eqs. (51). The transformed Eqs. (51) are greatly simplified by the condition $k'_{y'} = 0$.

For $k, s \rightarrow 0$, the solution of the transformed equations is very simple. We set s = 0, we let $\vec{k} + \vec{k}' \rightarrow \vec{k}'$ for all \vec{k}' , but we keep the linear factors of k which, when multiplied by the factor of k already present in the bilinear viscosity term in Eq. (56), will yield the k^2 factor in the damping coefficient, $k^2\eta(k, s)$. A little algebra now gives

$$\lim_{k \to 0} \langle [p_{k+k}^{x'} p_{-k}^{x'}](s=0) p_{-k}^{y} \rangle k^{\prime 2} (\gamma_{\parallel} - \epsilon_{x}^{x'} x_{x}^{*})$$
$$= \lim_{k \to 0} \{ i k_{x'} k_{B} T [1 + \frac{1}{2} (1 - f/g^{2})] \langle p_{k}^{x'}(s=0) p_{-k}^{y} \rangle$$

$$+ik_{y} k_{B}T^{\frac{1}{2}}(1-f/g^{2})\langle p_{k}^{y'}(s=0)p_{-k}^{y}\rangle \Big\} , \qquad (58)$$

$$\lim_{k \to 0} \left\langle \left[p_{k+k}^{\mathbf{y}^{\prime}} e^{\mathbf{p}_{k+k}^{\prime}} \right] (s=0) p_{-k}^{\mathbf{y}} \right\rangle k^{\prime 2} (\gamma_{\perp} - \epsilon_{\mathbf{x}^{\prime}}^{\mathbf{x}^{\prime}}) \\ = \lim_{k \to 0} \left[i k_{\mathbf{y}^{\prime}} k_{B} T \left\langle p_{k}^{\mathbf{y}^{\prime}} (s=0) p_{-k}^{\mathbf{y}} \right\rangle \right], \quad (59)$$

 $\lim_{k \to 0} \left\langle \left[p_{k+k}^{x^{\bullet}} p_{-k}^{y^{\bullet}} \right] (s=0) p_{-k}^{y} \right\rangle$

$$\times \left[k^{\prime 2}(\gamma_{\parallel} + \gamma_{\perp} - 2\epsilon_{y}^{*} \epsilon_{x}^{*}) + k_{B}T/mg\beta_{\perp}\right]$$
$$= \lim_{k \to 0} \left[ik_{x} \epsilon_{B}T \langle p_{k}^{y} (s=0)p_{-k}^{y} \rangle + ik_{y} \epsilon_{B}T \langle p_{k}^{*} (s=0)p_{-k}^{y} \rangle\right], \quad (60)$$

where the subscripts \parallel and \perp refer to the longitudinal and transverse components, respectively, of the bare viscosities α , β , and γ . The subscripts and superscripts on ϵ refer only to the indices for the stress tensor at time zero and time t, respectively, in Eq. (50); we have ignored the momentum indices. The next step is to perform the inverse transformations, expressing $p^{y'}$, $p^{x'}$, $k_{x'}$, and $k_{y'}$ in terms of p^x , p^y , and k. When we substitute the inverse-transformed Eqs. (58)-(60) into Eq. (56), we find that the p^x correlation functions are multiplied by angular functions which will vanish in the sum over k'; thus, we now drop those terms. The inverse-transformed Eqs. (58)-(60) yield

$$\lim_{k \to 0} \sin \phi \cos \phi \left\langle \left[p_{k+k}^{\mathbf{x}^{\prime}} \cdot p_{-k}^{\mathbf{y}^{\prime}} - p_{k+k}^{\mathbf{y}^{\prime}} \cdot p_{-k}^{\mathbf{y}^{\prime}} \right] (s=0) p_{-k}^{\mathbf{y}} \right\rangle$$

$$= \lim_{k \to 0} \frac{ikk_B T}{k'^2} \sin^2 \phi \cos^2 \phi \left(\frac{1}{\gamma_{\parallel} - \epsilon_{xx}^{xx}} + \frac{1}{\gamma_{\perp} - \epsilon_{xy}^{xy}} \right) \\ \times \langle p_k^y (s=0) p_{-k}^y \rangle, \quad (61)$$

 $\lim_{k \to 0} (\cos^2 \phi - \sin^2 \phi) \langle [p_{k+k^{\bullet}}^{x^{\bullet}} p_{-k^{\bullet}}^{y^{\bullet}} + p_{k+k^{\bullet}}^{y^{\bullet}} p_{-k^{\bullet}}^{x^{\bullet}}] (s = 0) p_{-k}^{y} \rangle$

$$= \lim_{k \to 0} ikk_B T(\cos^2\phi - \sin^2\phi)^2$$

$$\times \frac{1}{k'^2(\gamma_{\parallel}+\gamma_{\perp}-2\epsilon_{xy}^{xx})+k_BT/mg\beta_{\perp}} \langle p_k^y(s=0)p_{-k}^y \rangle.$$
(62)

Equations (61), (62), (57), and (56) may now be combined to complete the $k, s \rightarrow 0$ calculation. The sum $\sum_{k'=0}^{k}$ in Eq. (56) is replaced by

$$\frac{V}{(2\pi)^2} \int_0^{2\pi} d\phi \int_0^{k_c} k' \, dk' \; .$$

We now see that the k' integral in Eq. (56) posseses a logarithmic infinity due to the contribution to the integrand of the x'x' and y'y' terms in Eq. (61), whereas the contribution of the x'y' and y'x'terms in Eq. (62) is finite, and therefore negligible. Since $\vec{k}' = \vec{k} + \vec{k}'$ is parallel to x', $p^{x'}p^{x'}$ may be thought of as a pair of longitudinal fluctuations, $p^{y'}p^{y'}$ as a pair of transverse fluctuations, and $p^{x'}p^{y'}$ or $p^{y'}p^{x'}$ as a transverse fluctuation plus a longitudinal fluctuation. Thus, we have obtained the well known result that the major bilinear contributions to the transport coefficients (neglecting heat modes) come from pairs of transverse and pairs of longitudinal (sound) modes. This comparison of our results with simpler versions of the mode-mode theory is made possible by the transformation to the (x', y') frame. Our final equations are

$$\lim_{k \to 0} \langle p_k^{\mathbf{y}}(s=0) p_{-k}^{\mathbf{y}} \rangle \langle p_k^{\mathbf{y}} p_{-k}^{\mathbf{y}} \rangle^{-1} = \left[k^2 \alpha_{\perp} + \frac{k^2 k_B T}{16 \pi \rho} \right] \\ \times \left(\frac{1}{\gamma_{\parallel} - \epsilon_{xx}^{xx}} + \frac{1}{\gamma_{\perp} - \epsilon_{xy}^{xy}} \right) \int_0^{k_c} \frac{dk'}{k'} \right]^{-1} , \quad (63a)$$
$$\eta(k=0, \ s=0) = \alpha_{\perp} + \frac{k_B T}{16 \pi \rho} \left(\frac{1}{\gamma_{\parallel} - \epsilon_{xx}^{xx}} + \frac{1}{\gamma_{\perp} - \epsilon_{xy}^{xy}} \right) \\ \times \left\{ k_{\perp}^{k_c} \frac{dk'}{k'} \right\} , \quad (63b)$$

i.e., $\eta(k=0, s=0)=\infty$. It is actually inconsistent to retain α_{\perp} in Eqs. (63), as we have discarded bilinear contributions of possibly comparable magnitude. We retain terms like α_{\perp} , and shall continue to do so, for illustration.

Equations (63) contain no proof of the presence of t^{-1} tails on the flux autocorrelation function in

two dimensions. However, the infinite $\eta(k=0,$ s = 0) seen in Eq. (63b) implies that, for k = 0, the flux autocorrelation function [inverse Laplace transform of $\eta(k, s)$] must decay as t^{-1} , or more slowly, at long times; the logarithmic infinity in Eq. (63b) is very suggestive of a t^{-1} tail. One also sees from Eq. (63b) that it is not permissible to perform a k expansion on $\eta(k, s)$. These results have been obtained self-consistently. We have not assumed that a k expansion is valid to prove that it is not; we have assumed that a k, k' expansion is valid. We have not assumed that $\eta(k, s)$ is well behaved for small k and s; we have assumed this for α , β , and γ . The coefficients that appear in Eqs. (63), α , ϵ (which we expect to be small), and γ , do not appear at all in ordinary hydrodynamics. Furthermore, γ , which involves the flux autocorrelation function for p_{k+k}, p_{-k} [i.e., (1-P) $\times (d/dt) p_{k+k} p_{-k}$, is not trivially related to any definition of the viscosity in terms of the usual stress tensor [(1-P)p] correlation function.

B. Calculation of $\eta(0, s)$

We shall now present the results of the calculation of $\langle p_k^y(s) p_{-k}^y \rangle \cdot \langle p_k^y p_{-k}^y \rangle^{-1}$ for $k \to 0$ (i.e., we neglect k with respect to k') but for small finite s. This calculation is still rather simple, as the various bilinear momentum products, expressed in the (x', y') coordinate frame, still correspond to products of pure longitudinal and transverse modes; the algebra is correspondingly uncomplicated. Proceeding exactly as in Sec. IIIA, we obtain

$$\begin{split} \lim_{k \to 0} & \langle p_{k}^{y}(s) p_{-k}^{y} \rangle \langle p_{k}^{y} p_{-k}^{y} \rangle^{-1} = \left\{ s + k^{2} \alpha_{\perp} \right. \\ & + k^{2} \left. \frac{k_{B} T}{16 \pi \rho} \left. \frac{1}{2} \left[\left(\gamma_{\parallel} - \epsilon_{xx}^{xx} \right)^{-1} \ln \left(\frac{(\gamma_{\parallel} - \epsilon_{xx}^{xx}) k_{\sigma}^{2} + s}{s} \right) \right. \right. \\ & + \left(\gamma_{\perp} - \epsilon_{xy}^{xy} \right)^{-1} \ln \left(\frac{2(\gamma_{\perp} - \epsilon_{xy}^{xy}) k_{\sigma}^{2} + s}{s} \right) \right] \right\}^{-1} \quad (64a) \end{split}$$

and

$$\lim_{k \to 0} \eta(k, s) = \alpha_{\perp} + \frac{k_B T}{16\pi\rho} \frac{1}{2} \left[(\gamma_{\parallel} - \epsilon_{xx}^{xx})^{-1} \ln\left(\frac{(\gamma_{\parallel} - \epsilon_{xx}^{xx})k_c^2 + s}{s}\right) + (\gamma_{\perp} - \epsilon_{xy}^{xy})^{-1} \ln\left(\frac{2(\gamma_{\perp} - \epsilon_{xy}^{xy})k_c^2 + s}{s}\right) \right]. \quad (64b)$$

Equation (64b) explicitly contains the t^{-1} tail which we have sought to investigate. An inverse Laplace transform yields

$$\lim_{k \to 0} \langle j_k(t) j_{-k} \rangle^{\dagger} = 2 \alpha_{\perp} \delta(t)$$
$$+ \frac{k_B T}{32\pi\rho} \left(\frac{(\gamma_{\parallel} - \epsilon_{xx}^{xx})^{-1} (1 - e^{-k_c^2 (\gamma_{\parallel} - \epsilon_{xx}^{xx})t})}{t} \right)$$

+
$$\frac{(\gamma_{\perp} - \epsilon_{xy}^{xy})^{-1}(1 - e^{-2k_c^2(\gamma_{\perp} - \epsilon_{xy}^{xy})t})}{t}$$
, (65)

where j is the "flux" for momentum transport. For long times, and $k \rightarrow 0$ the projected flux autocorrelation function decays as t^{-1} . Note that the decaying exponentials for longitudinal and transverse modes in Eq. (65) have decay coefficients $k_c^2(\gamma_{\parallel}-\epsilon_{xx}^{xx})$ and $2k_c^2(\gamma_{\perp}-\epsilon_{xy}^{xx})$, respectively; this is a manifestation of the fact that the damping coefficients for longitudinal and transverse momentum fluctuations are proportional to $\frac{1}{2}(\gamma_{\parallel} - \epsilon_{xx}^{xx})$ and $(\gamma_{\perp} - \epsilon_{xy}^{xy})$, respectively. Equation (64b) is very similar to Eq. (59) of Zwanzig's paper (he treats the case $k \rightarrow 0$, finite s).^{2(b)} The important differences between the two equations arise because Zwanzig has considered an imcompressible fluid. and because his treatment involves a single bare viscosity ν , as opposed to our four bare viscosities ϵ , α , β , and γ .

C. Calculation of $\eta(k, 0)$

In the above sections, we have obtained results which are valid for $k \ll k'$. We have already pointed out, however, that one result of a proper treatment of the bilinear effects might be a breakdown of the ordinary small-k expansion for $\eta(k, s)$. Thus, it is of major interest to extend our calculations to the case of finite k. Furthermore, as we have discussed, ignoring k with respect to k'introduces a small error into the zero-k result, i.e., into our earlier calculations. Recall that we are assuming that a k, k' expansion of Eqs. (51) is valid even if a k expansion is not, and thus we may obtain, consistently, an expression for $\eta(k, s)$ which could never be obtained via a k expansion.

The algebra necessary to obtain $\langle p_k(s) p_{-k} \rangle$ $\langle p_k p_{-k} \rangle^{-1}$ from Eqs. (51) is greatly complicated for finite k. Since $\vec{k} + \vec{k'}$ is no longer parallel to $\mathbf{k}'(x')$, $p_{k+k}^{x'}$ and $p_{k+k'}^{y'}$ no longer correspond to pure longitudinal and transverse fluctuations; algebraically, the difficulties arise because $(\mathbf{k} + \mathbf{k}')^{\mathbf{y}'} \neq 0$. In order to make progress here we shall set γ_{μ} = $\gamma_1 = \gamma_2$, and similarly for β . We shall set $\epsilon = 0$. It should be clear from the results which we have already obtained that this approximation should not fundamentally affect the validity of what is to follow.

We now consider the case of finite k, and s equal to zero. Equations (51) yield

$$\lim_{s \to 0} \langle \dot{p}_{k}^{y}(s) \dot{p}_{-k}^{y} \rangle \langle \dot{p}_{k}^{y} \dot{p}_{-k}^{y} \rangle^{-1} = \left(k^{2} \alpha_{\perp} + \frac{k^{2} k_{B} T}{Nm} \frac{1}{2} \sum_{\mathbf{k}} (D_{\perp} + D_{\parallel}) \right)^{-1} \quad (66a)$$

and

s **→** 0

$$\lim_{s \to 0} \eta(k, s) = \alpha_{\perp} + \frac{k_B T}{Nm} \frac{1}{2} \sum_{\vec{k}} (D_{\perp} + D_{\parallel}), \quad (66b)$$

where

T. KEYES AND I. OPPENHEIM

$$D_{1} = \frac{\gamma(\kappa^{2} + k'^{2}) + \omega/k'^{2} [2k'\sin\phi\cos\phi + k\sin\phi]^{2}}{\gamma(\kappa^{2} + k'^{2}) [\gamma(\kappa^{2} + k'^{2}) + \omega\kappa^{2}/k'^{2}]},$$
(67a)

$$D_{\parallel} = \frac{\gamma(\kappa^{2} + k'^{2}) + \omega(k'^{2}/\kappa^{2}) + \omega([k\cos\phi + k'(\cos^{2}\phi - \sin^{2}\phi)]^{2} \left\{ \frac{1}{k'^{2}} - \frac{1}{2}k'^{2}[(1/\kappa^{2}) + (1/k'^{2})]^{2} \right\}}{\gamma^{2}(\kappa^{2} + k'^{2})^{2} + \left[\frac{1}{2}\gamma(\kappa^{2} + k'^{2})/\kappa^{2}k'^{2}\omega[3k'^{4} + \kappa^{4} - 2\kappa^{2}k'^{2}] + \frac{1}{2}\omega^{2}(1 - k'^{2}/\kappa^{2})^{2}} \right]}, \quad (67b)$$

$$\omega = k_B T / mg\beta , \qquad (68)$$

and $\kappa^2 = |\vec{k} + \vec{k}'|^2$. The quantities D_{\parallel} and D_{\perp} represent the bilinear effects due to pairs of longitudinal and transverse modes, respectively. It is easy to see that, as $k \to 0$ (recall that k' is small), both D_{\parallel} and D_{\perp} reduce to $2\sin^2\phi \cos^2\phi/\gamma k'^2$, in agreement with the results already obtained. It seems clear that γ should be set equal to γ_{\parallel} for D_{\parallel} , and to γ_{\perp} for D_{\perp} .

The integrals which arise in Eqs. (66), after the substitution

$$\sum_{\vec{k'}} - \frac{V}{(2\pi)^2} \int_0^{2\pi} d\phi \int_0^{k_c} dk' \, k'$$

are evaluated by first transforming the angular integral into a contour integral around the unit circle. We then divide the k' integral into three parts,

$$\int_{0}^{k_{c}} = \int_{0}^{Ak} + \int_{Ak}^{Bk} + \int_{Bk}^{k_{c}}, \qquad (69)$$

where A and B are numbers less than and greater than one, respectively. For the three regions of integration, we have $k' \ll k$, $k' \approx k$, and $k' \gg k$, respectively. Upon evaluating the integrations, we find, for small k,

$$\int_{0}^{k_{c}} \approx \int_{Bk}^{k_{c}}$$
(70)

The final result is

$$\lim_{s \to 0} \langle p_k^y(s) p_{-k}^y \rangle \langle p_k^y p_{-k}^y \rangle^{-1} = \left\{ k^2 \alpha_{\perp} + k^2 \frac{k_B T}{16 \pi \rho} \left[\frac{1}{\gamma_{\perp}} \ln \frac{k_c}{k} + \frac{1}{2} \frac{1}{\gamma_{\parallel}} \ln \frac{k_c}{k} \right] \right\}^{-1},$$
(71a)

 $\lim_{s \to 0} \eta(k, s)$

$$= \alpha_{\perp} + \frac{k_B T}{16 \pi \rho} \left[\frac{1}{\gamma_{\perp}} \ln\left(\frac{k_c}{k}\right) + \frac{1}{2} \frac{1}{\gamma_{\parallel}} \ln\left(\frac{k_c}{k}\right) \right] ,$$
(71b)

where we have replaced γ by the appropriate γ_{\parallel} or γ_{\perp} , as indicated above, and we have noted that, for small k/k_c , $\ln(k_c/Bk) \approx \ln(k_c/k)$.

For $k \rightarrow 0$, Eqs. (71) differ slightly from our k, $s \rightarrow 0$ results obtained earlier [Eqs. (63)], owing to the factor of $\frac{1}{2}$ multiplying the $1/\gamma_{\parallel}\ln(k_c/k)$ terms in Eqs. (71). This factor arises because, for k' > k, the third term in the denominator of D_{\parallel} must be kept, even though this term vanishes if k = 0. For finite \vec{k} , a product of sound modes with wave vectors $\vec{k} + \vec{k}'$ and $-\vec{k}'$ possesses an oscillatory tory part with frequency $\sim |\vec{k}|$; such oscillatory time behavior tends to destroy any divergences in the transport coefficients. This discussion indicates that one must be very careful in taking the $k \rightarrow 0$ limit, and that our earlier derivations have not been rigorously correct.

The most interesting aspect of Eqs. (71) is the presence of the $\ln(k_c/k)$ terms in $\eta(k, s)$. Clearly, a term of form $\ln(k_c/k)$ indicates that a k expansion of the dynamical laws is invalid in two dimensions; such a term could not be consistently obtained in any theory which assumes the k expansion. We have, however, obtained our results completely consistently via the k, k' expansion.

D. Calculation of $\eta(k, s)$

The most interesting calculation which we may perform, of course, is that for the case of finite, small k and s. In order to perform this calculation, in addition to our above assumption that $\gamma_{\parallel} = \gamma_{\perp} = \gamma$, $\beta_{\parallel} = \beta_{\perp} = \beta$, and $\epsilon = 0$ we shall make the less trivial assumption that the fluid is incompressible. This means that we set $n_k = 0$ in Eqs. (51). The effect of our incompressibility assumption is that we treat the sound (propagating) modes of the fluid as nonpropagating modes. Thus, we expect that our results shall overcount the bilinear contributions to $\eta(k, s)$ by a factor of 2; the true bilinear contribution from a sound mode plus a shear mode, which was negligible in the previous calculation [see discussion following Eq. (62)], shall be treated as a pair of shear modes, and will enter into our result. However, no important general aspects of the $t^{-d/2}$ behavior should depend upon the compressibility of the fluid, and we expect our results to be basically correct.

The solution of Eqs. (51) for $n_k = 0$, is easy;

$$\langle p_{k}^{y}(s) p_{-k}^{y} \rangle^{-1} \langle p_{k}^{y} p_{-k}^{y} \rangle^{-1}$$

$$= \left[s + k^{2} \alpha_{\perp} + \frac{k^{2} k_{B} T}{8 \pi \rho \gamma} \ln \left(\frac{s + 2k_{c}^{2} \gamma}{s + k^{2} \gamma} \right) \right]^{-1}, \quad (72a)$$

$$\eta(k, s) = \alpha_{\perp} + \frac{k_B T}{8\pi\rho\gamma} \ln\left(\frac{s + 2k_c^2\gamma}{s + k^2\gamma}\right) \quad .$$
(72b)

Comparison of Eqs. (72) with Eqs. (64) tends to confirm our earlier speculation that the effect of ignoring compressibility (besides eliminating the difference between γ_{\parallel} and γ_{\perp}) is to overcount the bilinear part of $\eta(k, s)$ by a factor of 2. In com-

paring our finite-s results with our zero-s results one should note that setting s to zero introduces a factor of 2, i.e., $\ln(s+k^2) \rightarrow 2\ln k$.

With Eqs. (72), which are very simple to derive via our method, we have obtained the full k and sdependence of the bilinear effects. The inverse Laplace transform of Eq. (72b) yields

$$\langle j_k(t)j_{-k}\rangle^{\dagger} = \alpha_{\perp}\delta(t) + \frac{k_B T}{8\pi\rho\gamma} \frac{e^{-k^2\gamma t} - e^{-2k_c^2\gamma t}}{t}.$$
 (73)

Thus, we see that the t^{-1} tail is present only as $k \to 0$. The "protection" of the t^{-1} tail for finite k, which also occurs for the three-dimensional $t^{-3/2}$ tail, will play an important role in any experimental attempt to observe the long-time tails.

IV. DISCUSSION

We now summarize the major points of the article. We have assumed that there exists a set of coupled transport equations for the correlation functions of the linear (L_k) and bilinear $(L_{k+k}, L'_{-k'})$ hydrodynamic variables, which possess wavevector- and frequency-independent transport coefficients. We have assumed that the bilinear equations may be expanded analytically up to quadratic order in k and k'. We have also assumed that we need only consider bilinear variables, $L_{k+k'}L'_{-k'}$, for k' less than the cutoff wave vector k_c ; k_c is considered sufficiently small such that the k, k' expansion of the dynamical laws may be consistently terminated at quadratic order. These assumptions constitute our "model."

The solution of the bilinear equations for the transverse momentum density autocorrelation functions yields, by comparison with a formally exact linear law, the k and s dependent shear viscosity coefficient, $\eta(k, s)$, of the true linear equations. We have seen that $\eta(k, s)$ has an important frequency dependence, and is nonanalytic in k; these results are completely consistent with our assumption of well-behaved bilinear equations. Thus, we feel that we have dealt with the self-consistency problem of the simpler theories^{2(a)} discussed in the Introduction.

It is our conclusion that, as $k \to 0$, the viscous flux-autocorrelation function indeed decays as t^{-1} in two dimensions, in agreement with all the other theories. One should note that our expressions

*Work supported in part by a grant from the National Science Foundation.

for the long-time part of the flux-autocorrelation function involve the bare transport coefficient γ which does not enter into ordinary hydrodynamics; this underscores the impossibility of obtaining a theory of the long-time tails via linear hydrodynamics. It is also important to note that the longtime tails exist only as k - 0; this consideration (which also applies to the three-dimensional $t^{-3/2}$ tails) must be taken into account when searching for experiments to observe the long-time phenomena.

It is of interest to consider the density dependence of $\eta(k, s)$ [Eq. (72b)]. Recall that we have always referred to "kinematic viscosities," so it is necessary to multiply our $\eta(k, s)$ by a factor of ρ to obtain the ordinary shear viscosity coefficient. Furthermore, it is easy to see that α and γ behave as $\rho^{-1}\alpha_0$ and $\rho^{-1}\gamma_0$, respectively, where α_0 and γ_0 are independent of ρ for small ρ . Thus, we may write the low density form of Eq. (72b) as

$$\rho\eta(k, s) = \alpha_0 + \frac{\rho k_B T}{8\pi\gamma_0} \ln\left(\frac{\rho s + 2k_c^2\gamma_0}{\rho s + k^2\gamma_0}\right).$$
(74)

Equation (74) shows that the bilinear contribution to the shear viscosity coefficient vanishes as the density approaches zero, in agreement with the low-density, kinetic-theory treatments⁹ of the bilinear effects. The bare transport coefficients are difficult to determine experimentally but could be obtained in principle if $\eta(k, s)$ were measured over a range of k and s values.

Much further work remains to be done on the nonlinear dynamics of collective modes. The first problem is to extend the treatment in Sec. III to three dimensions. We feel that a self-consistent treatment of triple and higher-order nonlinearities is of great importance. It is also desirable that the connection between our approach and the kinetic theories⁹ mentioned above be explored. Such an exploration would illuminate the interrelated roles of the density, the wave vector, and the order of nonlinearity as expansion parameters for the collective dynamical laws.

Finally, it is of interest to evaluate the consequences of the nonlinear effects upon the macroscopic hydrodynamic equations, the equations describing the decay to equilibrium of nonequilibrium averaged quantities. Work is currently in progress upon the above-mentioned topics.

[†]National Science Foundation Postdoctoral Fellow.

¹B. J. Alder and T. E. Wainwright, Phys. Rev. A 1, 18 (1970). ²(a)M. H. Ernst, E. H. Hauge, and J. M. J. van Leeuwen, Phys. Rev. Lett. 25, 1254 (1970); Ark. Fys. 8, (1971); Y. Pomeau, Phys. Rev. A 5, 2569 (1972). (b) Robert Zwanzig, Proceedings of the Sixth IUPAC Conference on Statistical Mechanics, edited by Stuart A. Rice, Karl F. Freed, and John C. Light (University of Chicago Press, Chicago, 1972).

³K. Kawasaki, Ann. Phys. (N.Y.) 61, 1 (1970).

⁴L. Landau and G. Placzeck, Phys. Z. Sowjetunion 5, 172 (1934).

⁵H. Mori, Prog. Theor. Phys. 33, 423 (1965).

⁶B. U. Felderhof and I. Oppenheim, Physica (The Hague) **31**, 1441 (1965).

⁷L. Onsager, Phys. Rev. 37, 405 (1931).

⁸J. H. Irving and J. G. Kirkwood, J. Chem. Phys. 18, 817 (1950). ⁹M. H. Ernst and J. R. Dorfman, Physica (The Hague) 61, 157 (1972); J. R. Dorfman and E. G. D. Cohen, Phys. Rev. Lett. 25, 1257 (1970); Y. Pomeau, Phys. Lett. A 27, 601 (1968); James W. Dufty, Phys. Rev. A 5, 2247 (1972); Gene F. Mazenko, report of work prior to publication.