Theory of Nonlinear Transport Processes: Nonlinear Shear Viscosity and Normal Stress Effects*

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Formally exact equations of motion satisfied by the gross variables of a macroscopic system valid far from thermal equilibrium are obtained with the aid of the new projection operator in nonequilibrium statistical mechanics. These equations are used to study nonlinear shear viscosity and normal stress effects of a model incompressible fluid in the presence of a shear flow which is not necessarily small. We find that the mode-coupling mechanism responsible for the long-time tails in time-correlation functions becomes very important here also and that a simple power-series expansion in the rate of shear D therefore fails. The part of shear viscosity dependent upon the rate of shear and the normal stress effect are found to vary as $|D|^{1/2}$ and $|D|^{3/2}$, respectively.

I. INTRODUCTION

During the past two decades or so we have witnessed tremendous advances in the statistical-mechanical theory of transport processes owing to the introduction of time-correlation functions of fluctuations, for situations for which the deviations from thermal equilibrium are small. The correctness of this new approach has been tested on a great number of examples, and this development is now well documented.[1] Recently, various ambitious attempts have been made to extend this new approach to the states not near thermal equilibrium.[2,3] In particular, Robertson[3] put forward rather general equations of motion of macroscopic variables employing the powerful projection-operator techniques originated by Zwanzig.[4] However, much (not all) of these attempts stop at a formal stage and their status in nonequilibrium statistical mechanics remains obscure.

In this paper, as well as in its sequel, we also derive first formally exact equations of change introducing a new projection operator which are valid arbitrarily far from thermal equilibrium, but furthermore apply the formalism to various model systems to obtain specific macroscopic equations of change.

We believe that the works along this line are also relevant for transport theory in general. Criticisms are still being raised against the correlation function approach to transport coefficients mentioned at the beginning, which we must take seriously.[5] However, in the linear regime, the results of the correlation function theory have been checked with the predictions of the Boltzmann equation for which these criticisms do not apply.[1] It is then conceivable that the difficulty, if any, may surface in the nonlinear regime.

As specific applications of the general formalism to be described in Sec. II we consider the nonlinear shear viscosity and the normal stress effects of a model incompressible fluid in Sec. III and Sec. IV. The motivation for our choice of this problem comes from the discovery in 1967 by Yamada and one of the authors[6] that in fact the so-called long-time tails of time-correlation functions[7,8] become very important in nonlinear regimes and are hence amenable to analysis by the mode-coupling theory. We shall find that indeed the nonlinear shear viscosity and normal stress effects can no longer be expressed as power-series expansions in the rate of shear, but contain fractional powers.

II. GENERAL THEORY OF NONLINEAR TRANSPORT

A. Local Equilibrium States

It is a common fact that a system of macroscopic size composed of a great number of microscopic constituents exhibits rather simple macroscopic behavior described in terms of a very small number of variables. Thus it is natural to expect that any theoretical attempts at a macroscopic description of such a system should start with identifying such variables which we shall call the gross variables. A generally accepted criterion is that if a system possesses a set of constants of the motion, the densities of these conserved quantities form the gross variables.[9] However, the choice is not free from arbitrariness. For instance, in the presence of a slow chemical reaction, concentrations of reacting components should be included in the gross variables although they are not conserved quantities. If the chosen set of the gross variables is inadequate, this fact manifests itself as a memory effect in the macroscopic equation of motion. In this section dealing with a general theory we henceforth assume the existence of such a set of gross variables to be denoted as $\{a\}$, whose ith member is a_i, and allow memory effects. For definiteness we consider an isolated system obeying classical mechanics which may be subjected to external forces. A state of the system is described by the nonequilibrium phase-space distribution function $\rho(t)$. Given a judicious choice of gross variables, a nonequilibrium state at time t can be specified to a good approximation by the values of the gross variables $\{a(t)\}$ at the same time. Namely, $\rho(t)$ can be approximated by the local equilibrium distribution function $\rho_l(t)$ whose time dependence comes solely from $\{a(t)\}$ and is given by[10]

$$\rho_l(t) = e^{\sigma_l(t)} \tag{2.1}$$

with

$$\sigma_l(t) \equiv -\Phi(\{b(t)\}) + \vec{A}\cdot\vec{b}(t), \tag{2.2}$$

where $\vec{A}$ is a vector whose ith component A_i is the phase-space function for the gross variable a_i and $\vec{b}(t)$ is another vector whose ith component $b_i(t)$ is the conjugate "field" of a_i and is determined so that the averages of $\vec{A}$ are identical for $\rho(t)$ and $\rho_l(t)$. Φ takes care of the normalization of $\rho_l(t)$. We borrow the notation of quantum mechanics and write Tr for phase-space integration, such that

$$\langle\vec{A}\rangle_t \equiv \mathrm{Tr}\vec{A}\rho(t) = \mathrm{Tr}\vec{A}\rho_l(t), \tag{2.3}$$

$$\mathrm{Tr}\rho(t) = \mathrm{Tr}\rho_l(t) = 1. \tag{2.4}$$

The difference between $\rho(t)$ and $\rho_l(t)$ is then responsible for irreversible and memory effects.[10] Near thermal equilibrium the problem has been studied a great deal.[1] In particular, use of the projection operator that extracts only the relevant part of $\rho(t)$ needed for macroscopic description has allowed rather concise treatments of the problem.[4] Here we extend this projection-operator formalism to the region far from equilibrium.

B. Projection Operators

We first introduce the following time-dependent projection operator $\mathcal{P}_l(t)$ acting upon an arbitrary phase-space function X:

$$\mathcal{P}_l(t)X \equiv \left(\rho_l(t) - \frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\langle\vec{A}\rangle_t\right)\mathrm{Tr}X + \frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\mathrm{Tr}\vec{A}X. \tag{2.5}$$

The time dependence of $\mathcal{P}_l(t)$ arises only through $\langle\vec{A}\rangle_t$. Then $\mathcal{P}_l(t)$ can be shown to have the following properties:

$$\mathcal{P}_l(t)\mathcal{P}_l(t') = \mathcal{P}_l(t), \tag{2.6a}$$

$$\mathcal{P}_l(t)\rho(t) = \rho_l(t), \tag{2.6b}$$

$$\mathcal{P}_l(t)\dot{\rho}(t) = \dot{\rho}_l(t), \tag{2.6c}$$

where a dot stands for time derivative. Note that $\mathcal{P}_l(t)\rho_l(t) = \rho_l(t)$ and $\mathcal{P}_l(t)1 \neq 1$. Note also that (2.6c) is a consequence of the fact that a small change in $\mathcal{P}_l(t)$ has no effect when operated upon $\rho(t)$, and hence if $\delta\rho$, $\delta\rho_l$ denote small variations, we have

$$\mathcal{P}_l(t)\,\delta\rho(t) = \delta\rho_l(t). \tag{2.6c$'$}$$

The property (2.6b) readily follows from the definition (2.5). The properties (2.6a) and (2.6c) can be also easily demonstrated (see Appendix A). The projection operator $\mathcal{P}_l(t)$ is similar to the one introduced by Robertson[3] except that he only retains the last term of (2.5), and hence the property (2.6b) is not satisfied. The property (2.6a) for $t = t'$ expresses the idempotent character of the projection operator. The properties (2.6b) and (2.6c) imply that $\mathcal{P}_l(t)$ picks up the part of the extremely complicated motion of $\rho(t)$ which varies very slowly through the succession of local equilibrium states. To see the connection with the more familiar definition of a projection operator,[11] we introduce another projection operator $\mathcal{P}_l(t)$ given by[12]

$$\mathcal{P}(t) \equiv \rho_l^{-1}(t)\mathcal{P}_l(t)\rho_l(t). \tag{2.7}$$

Then, for an arbitrary phase-space function X we readily find,

$$\mathcal{P}(t)X = \langle X\rangle_{lt} + \frac{\partial\sigma_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\langle\delta_t\vec{A}\,\delta_t X\rangle_{lt}, \tag{2.8}$$

where $\langle\cdots\rangle_{lt}$ denotes a local equilibrium average at time t and $\delta_t X \equiv X - \langle X\rangle_{lt}$, etc. Now, from the relation

$$\Phi(\{b\}) = \ln\mathrm{Tr}e^{\vec{A}\cdot\vec{b}} \tag{2.9}$$

together with (2.3) which relate $\vec{b}(t)$ and $\langle\vec{A}\rangle_t$ one finds

$$\frac{\partial\langle\vec{A}\rangle_t}{\partial\vec{b}(t)} = \langle\delta_t\vec{A}\,\delta_t\vec{A}\rangle_{lt} \tag{2.10}$$

and

$$\frac{\partial\sigma_l(t)}{\partial\vec{b}(t)} = \delta_t\vec{A}. \tag{2.11}$$

Therefore, using matrix notation, we obtain

$$\frac{\partial\sigma_l(t)}{\partial\langle\vec{A}\rangle_t} = \frac{\partial\sigma_l(t)}{\partial\vec{b}(t)}\cdot\frac{\partial\vec{b}(t)}{\partial\langle\vec{A}\rangle_t} = \delta_t\vec{A}\cdot\langle\delta_t\vec{A}\,\delta_t\vec{A}\rangle_{lt}^{-1}. \tag{2.12}$$

Finally (2.8) reduces to

$$\mathcal{P}(t)X = \langle X\rangle_{lt} + \delta_t\vec{A}\cdot\langle\delta_t\vec{A}\,\delta_t\vec{A}\rangle_{lt}^{-1}\cdot\langle\delta_t\vec{A}\,\delta_t X\rangle_{lt}\,. \tag{2.13}$$

If $\rho_l(t)$ in (2.13) is replaced by the equilibrium distribution function, $\mathcal{P}(t)$ indeed reduces to the classical-mechanical version of the projection operator introduced by Mori.[11] Therefore, our projection operator can be viewed as a rather natural extension of Mori's projection operator to the region far from equilibrium. In particular, $\mathcal{P}_l(t)$ picks up the slowly varying part of the distribution function $\rho(t)$ whereas $\mathcal{P}(t)$ picks up the slowly varying parts of dynamical variables.

C. Equations of Motion

Having introduced the projection operator, we make use of it to derive the exact equations of motion for $\langle\vec{A}\rangle_t$ which involve only the values of $\langle\vec{A}\rangle_s$ of the recent past and which reduce to the closed equations of motion for $\langle\vec{A}\rangle_t$ in the absence of memory effects.

We now start from the Liouville equation for $\rho(t)$,

$$\dot\rho(t) = -iL(t)\rho(t), \tag{2.14}$$

where the Liouville operator $L(t)$ may depend explicitly upon the time when time-dependent external forces are present. We transform (2.14) following a procedure similar to that employed by Zwanzig[4] and by Robertson.[3] First, using the property (2.6c) we have

$$\dot\rho_l(t) = -i\mathcal{P}_l(t)L(t)\rho_l(t) - i\mathcal{P}_l(t)L(t)[1-\mathcal{P}_l(t)]\rho(t). \tag{2.15}$$

The last term can be transformed by using

$$\frac{\partial}{\partial t}[1-\mathcal{P}_l(t)]\rho(t) = -i[1-\mathcal{P}_l(t)]L(t)\rho_l(t) - i[1-\mathcal{P}_l(t)]L(t)[1-\mathcal{P}_l(t)]\rho(t), \tag{2.16}$$

where we have made use of (A2). This can be integrated to yield

$$[1-\mathcal{P}_l(t)]\rho(t) = -\int_0^t ds\, U_l(ts)i[1-\mathcal{P}_l(s)]L(s)\rho_l(s) + U_l(t0)[1-\mathcal{P}_l(0)]\rho(0), \tag{2.17}$$

where we define

$$U_l(tt') = \begin{cases} \exp_+\left\{-i\int_{t'}^t ds[1-\mathcal{P}_l(s)]L(s)\right\}, & t\geq t', \\ \exp_-\left\{i\int_t^{t'} ds[1-\mathcal{P}_l(s)]L(s)\right\}, & t\leq t'. \end{cases} \tag{2.18}$$

Here $\exp_+$ and $\exp_-$ denote time-ordered exponentials in which operators are ordered from right to left and from left to right, respectively, as time increases. The last term of (2.17) represents the effect of the initial deviation from the local equilibrium state which is normally expected to disappear quickly. Thus we henceforth drop the last term of (2.17). The result is then substituted back into the last term of (2.15) to obtain

$$\dot\rho_l(t) = -i\mathcal{P}_l(t)L(t)\rho_l(t) - \int_0^t ds\,\mathcal{P}_l(t)L(t)U_l(ts) \times[1-\mathcal{P}_l(s)]L(s)\rho_l(s). \tag{2.19}$$

Here the first term represents the slow change of state through a succession of local equilibrium states whereas the second term contains irreversible processes arising from more random time variation. If we multiply (2.19) by A and integrate over the phase space, we obtain the equation of motion for $\langle\vec{A}\rangle_t$ which consists of two terms corresponding to the two terms of (2.19). We would like to have the second term of $\partial\langle\vec{A}\rangle_t/\partial t$ which contains irreversible processes expressed in the form of a time-correlation function of the random forces $[1-\mathcal{P}(s)]iL(s)\vec{A}$. For this purpose we further transform (2.19). First we note the following:

$$\frac{\partial}{\partial t}\langle\vec{A}\rangle_t = \mathrm{Tr}\vec{A}\,\dot\rho(t) = \mathrm{Tr}\vec{A}\,\dot\rho_l(t) = \langle\vec{A}\,\dot\sigma_l(t)\rangle_{lt}\,. \tag{2.20}$$

Therefore, we consider $\sigma_l(t)$ instead of $\rho_l(t)$ which by (2.19) obeys the following equation:

$$\dot\sigma_l(t) = -i\mathcal{P}(t)L(t)\sigma_l(t) - \int_0^t ds\,\mathcal{P}(t)\tilde L(t)\rho_l^{-1}(t)U_l(ts) \times\rho_l(s)[1-\mathcal{P}(s)]L(s)\sigma_l(s), \tag{2.21}$$

where

$$\tilde L(t) \equiv \rho_l^{-1}(t)L(t)\rho_l(t), \tag{2.22}$$

and we have used the fact that $L(t)$ is in fact a first-order differential operator. By differentiating and then integrating $\rho_l^{-1}(t)U_l(ts)\rho_l(s)$ with respect to t we readily find

$$\begin{aligned}\rho_l^{-1}(t)U_l(ts)\rho_l(s) &= \exp_+\left\{-\int_s^t ds'[\dot\sigma_l(s') + i(1-\mathcal{P}(s'))\tilde L(s')]\right\} \\ &= \exp_+\left[-\int_s^t R(ts')ds'\right]U(ts),\end{aligned} \tag{2.23}$$

where

$$R(ts) \equiv U(ts)\{[1-\mathcal{P}(s)][iL(s)\vec{A}]\cdot\vec{b}(s) + \delta_s\vec{A}\cdot\dot{\vec{b}}(s)\}U(st), \tag{2.24}$$

$$U(tt') \equiv \begin{cases} \exp_+\left\{-i\int_{t'}^{t} ds\,[1-\mathcal{P}(s)]L(s)\right\}, & t \geq t' \\ \exp_-\left\{i\int_{t}^{t'} ds\,[1-\mathcal{P}(s)]L(s)\right\}, & t \leq t'. \end{cases} \tag{2.25}$$

In deriving the last expression of (2.23) we made use of the fact that

$$\dot{\sigma}_l(t) = \delta_t \vec{A}\cdot\dot{\vec{b}}(t) \tag{2.26}$$

and the formula

$$\exp_+\left\{\int_s^t ds'[A(s')+B(s')]\right\} = \exp_+\left\{\int_s^t ds' \exp_+\left[\int_{s'}^t ds'' A(s'')\right] B(s') \times \exp_-\left[-\int_{s'}^t ds'' A(s'')\right]\right\} \exp_+\left[\int_s^t ds' A(s')\right] \tag{2.27}$$

which is valid for arbitrary operators $A(t)$ and $B(t)$.

The second term of (2.21) can be further transformed by noting that

$$\mathcal{P}_l(t)U_l(ts)[1-\mathcal{P}_l(s)] = 0 \tag{2.28}$$

which is a consequence of (2.6a). This allows us to insert $[1-\mathcal{P}_l(t)]$ in front of $U_l(ts)$ or $[1-\mathcal{P}(t)]$ just after $\tilde{L}(t)$ in (2.21). In this way we obtain

$$\dot{\sigma}_l(t) = -i\mathcal{P}(t)L(t)\sigma_l(t) - \int_0^t ds\,\mathcal{P}(t)\tilde{L}(t)[1-\mathcal{P}(t)] \times \exp_+\left[-\int_s^t R(ts')ds'\right] \times U(ts)[1-\mathcal{P}(s)]L(s)\sigma_l(s). \tag{2.29}$$

In this equation all the unknowns are expressed in terms of σ_l if $L(t)$ is given. Thus this is a closed equation for $\sigma_l(t)$ which depends only on the past history of σ_l. From this the equation for $\langle\vec{A}\rangle_t$ is readily obtained by substituting $\dot{\sigma}_l(t)$ into (2.20). Here we use the following properties:

$$\langle X\mathcal{P}(t)Y\rangle_{lt} = \langle Y\mathcal{P}(t)X\rangle_{lt}, \tag{2.30}$$

$$\langle X\tilde{L}(t)Y\rangle_{lt} = -\langle YL(t)X\rangle_{lt}. \tag{2.31}$$

Then we finally obtain

$$\frac{\partial}{\partial t}\langle\vec{A}\rangle_t = \langle iL(t)\vec{A}\rangle_{lt} - \int_0^t ds\left\langle\{[1-\mathcal{P}(t)]iL(t)\vec{A}\}\exp_+\left[-\int_s^t R(ts')ds'\right]U(ts)[1-\mathcal{P}(s)]iL(s)\vec{A}\right\rangle_{lt}\cdot\dot{\vec{b}}(s). \tag{2.32}$$

One can also proceed directly from (2.19) in which the second term of (2.19) can be written using (2.28) as

$$\int_0^t ds\,\mathcal{P}_l(t)L(t)[1-\mathcal{P}_l(t)]U_l(ts)[1-\mathcal{P}_l(s)]L(s)\rho_l(s). \tag{2.33}$$

After some algebra which we omit, we find

$$\frac{\partial}{\partial t}\langle\vec{A}\rangle_t = \langle iL(t)\vec{A}\rangle_{lt} - \int_0^t ds\,\langle\{[1-\mathcal{P}(t)][iL(t)\vec{A}]\}\rho_l^{-1}(t)U_l(ts)\rho_l(s)[1-\mathcal{P}(s)][iL(s)\vec{A}]\rangle_{lt}\cdot\dot{\vec{b}}(s) \tag{2.34}$$

which leads to (2.32) with the use of (2.23). In this manner we find that $\partial\langle A\rangle_t/\partial t$ consists of two terms, one which arises from a reversible change through a succession of local equilibrium states and another which arises from the random force $[1-\mathcal{P}(s)][iL(s)\vec{A}]$ and which is expressed as a time-correlation function of random force. Here $\rho_l^{-1}(t)U(ts)\rho_l(t)$ given by (2.23) describes the time development of random forces where the presence of the projection operator $[1-\mathcal{P}(s)]$ effectively eliminates long memory effects.[13] If we linearize with respect to deviations from thermal equilibrium, (2.32) is seen to reduce to the familiar expression of the linear response theory.[1] The equation similar to (2.32) with a somewhat different projection operator was obtained by Robertson.[3]

We now give an alternative expression of (2.32) which is suited to discuss steady states. First rewrite the integrand of the second term of (2.32) as

$$\left\langle\{[1-\mathcal{P}(t)]iL(t)\vec{A}\}\left(\frac{\partial}{\partial s}\exp_+\left[-\int_s^t R(ts')ds'\right] - \exp_+\left[-\int_s^t R(ts')ds'\right]\times U(ts)\,\delta_s\vec{A}\cdot\dot{\vec{b}}(s)\right)\right\rangle_{lt}, \tag{2.35}$$

where we understand that $\langle X\rangle_{lt} = \mathrm{Tr}\rho_l(t)(X\cdot 1)$ for any operator X, and where we have made use of the fact that $U(ts)\cdot 1 = 1$. Integrating this with respect to s we obtain

$$\frac{\partial}{\partial t}\langle \vec{A}\rangle_t - \langle iL(t)\vec{A}\rangle_{lt}$$
$$= \left\langle \{[1-\mathcal{P}(t)]iL(t)\vec{A}\}\left\{\exp_+\left[-\int_0^t R(ts)ds\right]-1 + \int_0^t ds\,\exp_+\left[-\int_s^t R(ts')ds'\right]U(ts)\,\delta_s\vec{A}\cdot\dot{\vec{b}}(s)\right\}\right\rangle_{lt}. \qquad (2.36)$$

The left-hand side is also written as $\mathrm{Tr}\rho(t)\vec{I}(t)$, where

$$\vec{I}(t) \equiv [1-\mathcal{P}(t)]iL(t)\vec{A} \qquad (2.37)$$

is the phase-space function for the irreversible current associated with $\vec{A}$ (which is in fact identical to the random force referred to above).

In a steady state where $\mathcal{P}$, L, and $\vec{b}$ no longer depend upon time, we have

$$R(ts) = e^{-i(t-s)(1-\mathcal{P})L}(1-\mathcal{P})(iL\vec{A})\,e^{i(t-s)(1-\mathcal{P})L}\cdot\vec{b}$$
$$\equiv R(t-s) \qquad (2.38)$$

which has a short correlation time. Thus the time integral of $R(ts)$ in (2.36) can be extended to $t\to\infty$, and we obtain for the average irreversible current $\langle I\rangle$ the following:

$$\langle I\rangle = \left\langle I\left\{\exp_-\left[-\int_0^\infty R(t)dt\right]-1\right\}\right\rangle_l, \qquad (2.39)$$

where the right-hand side is the average over the local equilibrium state corresponding to the steady state.

A special case of (2.39) has been obtained earlier for studying the nonlinear shear viscosity of fluids.[6] (See also Sec. III.) It would be of some interest to discuss the connection of the present approach to that given in Appendix A of Ref. 6. For this purpose we write

$$\rho(t) = \exp\sigma(t). \qquad (2.40)$$

Then (2.6c) can be written using (2.7) as

$$\tilde{\mathcal{P}}(t)\dot{\sigma}(t) = -\tilde{\mathcal{P}}(t)iL(t)\sigma(t) = \dot{\sigma}_l(t) \qquad (2.41)$$

with

$$\tilde{\mathcal{P}}(t) \equiv \rho_l^{-1}(t)\mathcal{P}_l(t)\rho(t) = \mathcal{P}(t)\,e^{\sigma'(t)} \qquad (2.42)$$

and

$$\sigma(t) = \sigma_l(t) + \sigma'(t). \qquad (2.43)$$

Here $\sigma'(t)$ represents the deviation from local equilibrium. Then, with $\dot{\sigma}'(t) = -iL(t)\sigma(t)$ we obtain

$$\dot{\sigma}'(t) = -i[1-\tilde{\mathcal{P}}(t)]L(t)[\sigma'(t)+\sigma_l(t)]. \qquad (2.44)$$

This is solved to yield

$$\sigma'(t) = -\int_0^t ds\,\exp_+\left\{-i\int_s^t ds'[1-\tilde{\mathcal{P}}(s')]L(s')\right\} \times i[1-\tilde{\mathcal{P}}(s)]L(s)\sigma_l(s) + \exp_+\left\{-i\int_0^t ds[1-\tilde{\mathcal{P}}(s)]L(s)\right\}\sigma'(0). \qquad (2.45)$$

This expression for $\sigma'(t)$ is only implicit because the right-hand side contains $\sigma'(t)$ through $\tilde{\mathcal{P}}(t)$, (2.42). If this $\sigma'(t)$ on the right-hand side of (2.42) is dropped, the result essentially reduces to that of Appendix A of Ref. 6 if $\sigma'(0)$ is also dropped and the stationarity condition is imposed. In general $\sigma'(t)$ can be ignored only if the system is close to the thermal equilibrium and, therefore, the approach adopted in this paper is more precise in the nonlinear domain. Strictly speaking $\mathcal{P}(t)\sigma(t)$ is not equal to $\sigma_l(t)$ as was assumed in Ref. 6.

III. MICROSCOPIC EXPRESSIONS FOR THE STEADY-STATE AVERAGE OF THE STRESS TENSOR IN FLUIDS

In Secs. III and IV we illustrate the use of the general theory of the preceding discussion by studying nonlinear shear viscosity and normal stress effects in one component fluids. To derive formal expressions for these effects we consider a fluid in a steady state in the presence of a uniform and constant rate of shear

$$D = \frac{\partial v_y}{\partial x}, \qquad (3.1)$$

where v_y is the y component of the average fluid velocity. We suppose that all other macroscopic variables such as v_x, v_z, temperature, and pressure assume their equilibrium values. The presence of the rate of shear then causes the average stress tensor in fluids, $P^{\alpha\beta} = P^{\beta\alpha}$, to deviate from its equilibrium value $p\delta_{\alpha\beta}$, where p is the equilibrium pressure. For isotropic fluids, considerations of reflection symmetries dictate the dependence of $P^{\alpha\beta}$ upon D up to some function of $|D|$. Namely, P^{zx} and P^{zy} must vanish whereas P^{xy} is an odd function of D, and $P^{\alpha\alpha}(\alpha = x, y, z)$ are even functions of D. The nonlinear shear viscosity $\eta(D)$ can then be defined as

$$P^{xy}(D) = -\eta(D)D \qquad (3.2)$$

and $\eta(0)$ is then the ordinary shear viscosity. The normal stress effects describe anisotropy in the diagonal components of the stress tensor which are typical nonlinear effects and are described by the differences $P^{xx}(D) - P^{yy}(D)$, etc.[14]

Let us now derive microscopic expressions for $P^{\alpha\beta}(D)$ where (2.39) and (2.38) can be used. We

TABLE I. Gross variables and their conjugate fields for one component fluid.

A_i	b_i
H	$-\beta$
$j^{\alpha}(\vec{r})$	$\beta v^{\alpha}(\vec{r})$
$\delta\rho(\vec{r})$	$\beta\mu(\vec{r})$

first construct the local equilibrium distribution function ρ_l which in this case can be readily obtained from the equilibrium distribution function ρ_0 merely by replacing all $\vec{p}_i$ by $\vec{p}_i - m\vec{v}(\vec{r}_i)$, m being the molecular mass. Therefore, we have

$$\rho_l = \rho_0 \exp\left[\beta \int \vec{j}(\vec{r})\cdot\vec{v}(\vec{r})d\vec{r} + \beta m \int \mu(\vec{r})\,\delta n(\vec{r})d\vec{r}\right] \tag{3.3}$$

with $\beta = 1/k_B T$ and

$$\mu(r) = -\tfrac{1}{2}[v(\vec{r})]^2, \tag{3.4}$$

$$\vec{j}(\vec{r}) \equiv \sum_i \vec{p}_i\,\delta(\vec{r}-\vec{r}_i) \quad \text{(mass current density)}, \tag{3.5}$$

where $\vec{r}_i$ and $\vec{p}_i$ are the coordinate and momentum of the ith molecule, and $\delta n(\vec{r})$ is the molecular expression for the number density minus its equilibrium average value. Equation (3.3) has the form of (2.1) and (2.2), where the gross variables $\vec{A}$ and their conjugate fields that enter ρ_l are given in the Table I, where H is the system Hamiltonian, and $j^{\alpha}(\vec{r})$ and $\delta n(\vec{r})$ form continuous density variables. The quantity $\mu(\vec{r})$ can be regarded also as a nonuniform chemical potential needed to keep the average chemical potential equal to its equilibrium value in the presence of the nonuniform velocity field. We note here that the complete set of the gross variables should include the Hamiltonian density instead of the total Hamiltonian, and this complete set must be used in defining $\mathcal{P}(t)$, (2.13).

Next we turn to the irreversible current I. Since H is a constant of the motion and $m\dot{n}(\vec{r}) = -\vec{\nabla}\cdot\vec{j}(\vec{r})$ is again a gross variable, the only irreversible current in the steady state under consideration is contained in $iLj^{\alpha}(\vec{r})$ whose molecular expression is

$$iLj^{\alpha}(\vec{r}) = -\sum_{\beta}\left(\frac{\partial}{\partial r_{\beta}}\right) J^{\alpha\beta}(\vec{r}), \tag{3.6}$$

where $J^{\alpha\beta}(\vec{r})$ is the molecular expression for the stress tensor given by

$$J^{\alpha\beta}(\vec{r}) = \frac{1}{m}\sum_i p_i^{\alpha} p_i^{\beta}\,\delta(\vec{r}-\vec{r}_i) - \frac{1}{2}\sum_{ij}(r_i^{\alpha}-r_j^{\alpha})\frac{\partial\phi_{ij}}{\partial r_i^{\beta}}\,\delta(\vec{r}-\vec{r}_i). \tag{3.7}$$

Here ϕ_{ij} is the intermolecular potential which is assumed to be centrally symmetric.

Although $\mathcal{P}J^{\alpha\beta}(\vec{r})$ can be directly obtained from (2.13), in this case there is an alternative easier way. For this purpose note that the local equilibrium distribution function (3.3) is obtained from ρ_0 simply by replacing all $\vec{p}_i$ by $\vec{p}_i - m\vec{v}(\vec{r}_i)$. Hence we introduce the following pseudocanonical transformation[6]:

$$S\vec{p}_i = \vec{p}_i - m\vec{v}(\vec{r}_i), \tag{3.8}$$

$$S\vec{r}_i = \vec{r}_i, \tag{3.9}$$

and then we have

$$\rho_l = S\rho_0. \tag{3.10}$$

The local equilibrium average of any phase space function X can now be written as

$$\langle X\rangle_l = \mathrm{Tr}X\rho_l = \mathrm{Tr}XS\rho_0 = \mathrm{Tr}\rho_0 S^{-1}X = \langle S^{-1}X\rangle_0, \tag{3.11}$$

where $\langle\cdots\rangle_0$ is the equilibrium average. The definition of the projection operator $\mathcal{P}$, (2.13), for steady states then becomes

$$\mathcal{P}X = \langle S^{-1}X\rangle_0 + S\delta(S^{-1}A)\cdot\langle\delta(S^{-1}A)\,\delta(S^{-1}A)\rangle_0^{-1}\cdot\langle\delta(S^{-1}A)\,\delta(S^{-1}X)\rangle_0, \tag{3.12}$$

where $\delta X \equiv X - \langle X\rangle_0$, etc., and we have used (3.11) together with the following:

$$\delta_l X = X - \langle X\rangle_l = X - \langle S^{-1}X\rangle_0 = S\delta(S^{-1}X) \tag{3.13}$$

for any phase-space function X, since the result of S operating upon a constant number is 1. On the other hand, $S^{-1}A_i$ can be written as a linear combination of the A's; for instance, $S^{-1}\vec{j}(\vec{r}) = \vec{j}(\vec{r}) - mn(\vec{r})\vec{v}(\vec{r})$. Then, since the definition of a projection operator does not depend upon a particular choice of the A's as long as they are equivalent sets of the gross variables, we can replace $S^{-1}A$ in (3.12) by A, and (3.12) finally reduces to

$$\mathcal{P}X = S[\langle S^{-1}X\rangle_0 + \delta A\cdot\langle\delta A\,\delta A\rangle_0^{-1}\cdot\langle\delta A\,\delta(S^{-1}X)\rangle_0] = S\mathcal{P}_0 S^{-1}X, \tag{3.14}$$

where $\mathcal{P}_0$ is the projection operator originally introduced by Mori[11];

$$\mathcal{P}_0 X = \langle X\rangle_0 + \delta A\cdot\langle\delta A\,\delta A\rangle_0^{-1}\cdot\langle\delta A\,\delta X\rangle_0. \tag{3.15}$$

With these preparations we obtain with (3.6)

$$(1-\mathcal{P})iL\vec{\mathrm{j}}=S(1-\mathcal{P}_0)S^{-1}iL\vec{\mathrm{j}}$$
$$=-\vec{\nabla}\cdot S(1-\mathcal{P}_0)S^{-1}\vec{\vec{\mathrm{J}}}=-\vec{\nabla}\cdot S(1-\mathcal{P}_0)\vec{\vec{\mathrm{J}}}, \qquad (3.16)$$

where we have noted that the difference $S^{-1}\vec{\vec{J}}-\vec{\vec{J}}$ is a linear combination of the A's and disappears by applying $(1-\mathcal{P}_0)$. The expression $(1-\mathcal{P}_0)\vec{\vec{J}}(\vec{r})$ was evaluated by Mori[11] with the result,

$$(1-\mathcal{P}_0)\vec{\vec{J}}(\vec{r})=\vec{\vec{J}}(\vec{r})-\left[p+\left(\frac{\partial p}{\partial e}\right)_n\delta H(\vec{r})+\left(\frac{\partial p}{\partial n}\right)_e\delta n(\vec{r})\right]\vec{\vec{1}}, \qquad (3.17)$$

where e is the average energy density, $\delta H(\vec{r})$ is the local Hamiltonian density minus e, and $\vec{\vec{1}}$ is the unit dyadic. Again using the fact that $S\vec{\vec{J}}(\vec{r})-\vec{\vec{J}}(\vec{r})$ is the linear combination of the δA's and $S\delta A$ is a linear combination of the δA's, we find

$$(1-\mathcal{P})iL\vec{\mathrm{j}}=-\vec{\nabla}\cdot\vec{\vec{J}}^*+(\text{the linear combination of the }\delta A\text{'s}) \qquad (3.18)$$

with

$$\vec{\vec{J}}^*\equiv\vec{\vec{J}}-\vec{v}\vec{\mathrm{j}}-\vec{\mathrm{j}}\vec{v}+mn\vec{v}\vec{v}-p\vec{\vec{1}}. \qquad (3.19)$$

It is also easy to show that

$$\left\langle A_i\left\{\exp_-\left[-\int_0^\infty R(t)dt\right]-1\right\}\right\rangle_l=0. \qquad (3.20)$$

This follows from the fact that by (2.38) this takes the form $\langle A_i(1-\mathcal{P})\cdots\rangle_l$ and

$$\langle\vec{A}(1-\mathcal{P})\cdots\rangle_l=\langle AS(1-\mathcal{P}_0)S^{-1}\cdots\rangle_l$$
$$=\langle S^{-1}AS(1-\mathcal{P}_0)S^{-1}\cdots\rangle_0$$
$$=\langle(S^{-1}A)(1-\mathcal{P}_0)S^{-1}\cdots\rangle_0=0, \qquad (3.21)$$

where we have used (3.11) and the fact that $S^{-1}\delta A$ is a linear combination of the δA's. The left-hand side of (2.39) can be written as $-\vec{\nabla}\cdot(\vec{\vec{P}}-\langle\vec{\vec{J}}\rangle_l)$. Then using (3.11) and

$$S^{-1}\vec{\vec{J}}(\vec{r})=\vec{\vec{J}}(\vec{r})+\vec{\mathrm{j}}(\vec{r})\vec{v}(\vec{r})+\vec{v}(\vec{r})\vec{\mathrm{j}}(\vec{r})+mn(\vec{r})\vec{v}(\vec{r})\vec{v}(\vec{r}) \qquad (3.22)$$

and also $\langle\vec{\vec{J}}(\vec{r})\rangle_0=p\vec{\vec{1}}$, we find the left-hand side is simply

$$\langle I\rangle=-\vec{\nabla}\cdot\{\vec{\vec{P}}-mn_0\vec{v}\vec{v}-p\vec{\vec{1}}\}=-\vec{\nabla}\cdot\vec{\vec{P}}^*, \qquad (3.23)$$

where n_0 is the equilibrium number density and

$$\vec{\vec{P}}^*=\vec{\vec{P}}-mn_0\vec{v}\vec{v}-p\vec{\vec{1}}=\langle\vec{\vec{J}}^*\rangle \qquad (3.24)$$

is precisely that part of the stress tensor due to irreversible processes. In this way we are led to the following expression for $\vec{\vec{P}}^*$:

$$\vec{\vec{P}}^*=\left\langle\vec{\vec{J}}^*\left\{\exp_-\left[-\int_0^\infty R(t)dt\right]-1\right\}\right\rangle_l, \qquad (3.25)$$

apart from an arbitrary contribution to $\vec{\vec{P}}^*$ whose divergence disappears, which can be ignored since it does not affect the hydrodynamic equations of motion of isotropic fluids. The expression for $R(t)$ is obtained after integration by parts as

$$R(t)=\beta\int d\vec{r}\,(\vec{\nabla}\vec{v}):e^{-it(1-\mathcal{P})L}(1-\mathcal{P})\vec{\vec{J}}\,e^{it(1-\mathcal{P})L}.$$

It is now possible to express (3.25) as the average over the equilibrium state. For this purpose note the following:

$$\vec{\vec{J}}^*=S(\vec{\vec{J}}-p\vec{\vec{1}}), \qquad (3.26)$$

$$R(t)=\beta\int d\vec{r}\,(\vec{\nabla}\vec{v}):\exp[-itS(1-\mathcal{P}_0)\tilde{L}S^{-1}]\times S(1-\mathcal{P}_0)S^{-1}\vec{\vec{J}}\exp[itS(1-\mathcal{P}_0)\tilde{L}S^{-1}]$$
$$=S\beta\int d\vec{r}\,(\vec{\nabla}\vec{v}):\exp[-it(1-\mathcal{P}_0)\tilde{L}]\times(1-\mathcal{P}_0)\vec{\vec{J}}'\exp[it(1-\mathcal{P}_0)\tilde{L}]S^{-1}, \qquad (3.27)$$

where

$$\tilde{L}\equiv S^{-1}LS,\quad \vec{\vec{J}}'\equiv S^{-1}\vec{\vec{J}}S. \qquad (3.28)$$

Hence using (3.11) we finally find the following exact expression for $\vec{\vec{P}}^*$:

$$\vec{\vec{P}}^*=\left\langle(\vec{\vec{J}}-p\vec{\vec{1}})\left\{\exp_-\left[-\int_0^\infty\tilde{R}(t)dt\right]-1\right\}\right\rangle_0 \qquad (3.29)$$

with

$$\tilde{R}(t)\equiv\beta\int d\vec{r}\,(\vec{\nabla}\vec{v}):e^{-it(1-\mathcal{P}_0)\tilde{L}}\times(1-\mathcal{P}_0)\vec{\vec{J}}'e^{it(1-\mathcal{P}_0)\tilde{L}}$$
$$=\beta D\int d\vec{r}\,e^{-it(1-\mathcal{P}_0)\tilde{L}}\times(1-\mathcal{P}_0)J'^{xy}e^{it(1-\mathcal{P}_0)\tilde{L}}. \qquad (3.30)$$

The expression for $\tilde{L}$ is also readily obtained as (see Appendix B of Ref. 6)[15]

$$\tilde{L}=L+DL', \qquad (3.31)$$

where

$$iL'\equiv\sum_i\left(x_i\frac{\partial}{\partial y_i}-p_i^x\frac{\partial}{\partial p_i^y}\right). \qquad (3.32)$$

In view of (3.2) we find the following exact expressions for the nonlinear shear viscosity:

$$\eta(D)=\frac{1}{D}\left\langle J^{xy}\left\{1-\exp_-\left[-\int_0^\infty\tilde{R}(t)dt\right]\right\}\right\rangle_0. \qquad (3.33)$$

The normal stress effects are likewise expressed as

$$P^{*xx}(D)-P^{*yy}(D)=\left\langle [J^{xx}-J^{yy}]\left\{\exp_-\left[-\int_0^\infty \tilde{R}(t)dt\right]-1\right\}\right\rangle_0 , \quad \text{etc.} \tag{3.34}$$

Before leaving this section we would like to make three remarks. First, in the thermodynamic limit ρ_0 as well as $\int J' d\vec{r}$ are invariant under spatial translation [see (3.39) below]; so is $\tilde{L}$ operated upon a translation invariant quantity. This allows us to limit the gross variables $\vec{A}$ appearing in the definition of the projection operator (3.15) to those having a zero wave vector in (3.30). Furthermore, we can replace $J^{\alpha\beta}(\vec{r})$ appearing in (3.33) and (3.34) by their spatial averages;

$$J^{\alpha\beta}(\vec{r})\to\frac{1}{V}I^{\alpha 3}=\frac{1}{V}\int J^{\alpha\beta}(\vec{r})d\vec{r}.$$

The second point comes from the fact that

$$\left\langle \exp_-\left[-\int_0^\infty \tilde{R}(t)dt\right]\right\rangle_0=1 \tag{3.35}$$

which is a consequence of the presence of $(1-\mathcal{P}_0)$ in $\tilde{R}(t)$. Equation (3.33) is then written as

$$\eta(D)=-\frac{1}{D}\frac{\langle J^{xy}\exp_-[-\int_0^\infty \tilde{R}(t)dt]\rangle_0}{\langle \exp_-[-\int_0^\infty \tilde{R}(t)dt]\rangle_0}. \tag{3.36}$$

Then it is well known that the formal perturbation expansion in powers of $\tilde{R}$ of this expression is obtained by replacing $\langle\cdots\rangle_0$ in the numerator by the cumulant average $\langle\cdots\rangle_{0c}$ where in each term of the expansion in $\tilde{R}$ of the form $\langle J^{xy}\tilde{R}(t_1)\tilde{R}(t_2)\cdots\tilde{R}(t_n)\rangle_{0c}$ all the factors J^{xy}, $\tilde{R}(t_1),\ldots,$ and $\tilde{R}(t_n)$ are simultaneously correlated among themselves, and then dropping the denominator of (3.36). This allows us to replace $\langle\cdots\rangle_0$ in (3.33) by $\langle\cdots\rangle_{0c}$. This is also true for (3.34). This result follows from the fact that the average in which some of the factors are uncorrelated necessarily contains at least a factor like $\langle\tilde{R}(t_1)\tilde{R}(t_2)\cdots\tilde{R}(t_m)\rangle_0$ which vanishes. As a final remark we show that for incompressible fluids J'^{xy} in (3.30) can be replaced by J^{xy}. To see this we note the following which results from the fact that at the boundary Σ the mass current $\vec{j}$ has zero normal component[16]:

$$0=\int_\Sigma d\vec{S}\cdot\vec{j}(\vec{r})g(\vec{r})=\int d\vec{r}\,\vec{\nabla}\cdot[g(\vec{r})\vec{j}(\vec{r})]=\int d\vec{r}\,\vec{j}(\vec{r})\cdot\vec{\nabla}g(\vec{r})-\frac{d}{dt}\int g(\vec{r})mn(\vec{r}t)d\vec{r}, \tag{3.37}$$

where $g(\vec{r})$ is an arbitrary function and we have used the continuity equation. Choosing $g(\vec{r})=x^\alpha x^\beta$ we have

$$\int d\vec{r}[x^\alpha j^\beta(\vec{r})+x^\beta j^\alpha(\vec{r})]=\frac{d}{dt}\int x^\alpha x^\beta mn(\vec{r}t)d\vec{r}. \tag{3.38}$$

On the other hand, we find

$$\vec{J}'=\vec{J}+\vec{v}\,\vec{j}+\vec{j}\,\vec{v}+mn\vec{v}\vec{v} \quad\text{or}\quad J'^{xy}=J^{xy}+Dxj^x. \tag{3.39}$$

Hence, we have

$$\int J'^{xy}(\vec{r})d\vec{r}=\int J^{xy}(\vec{r})d\vec{r}+D\int xj^x d\vec{r}, \tag{3.40}$$

where the second term is equal to $\frac{1}{2}mD\int x^2\dot{n}\,d\vec{r}$ which vanishes for an incompressible fluid which is the model fluid to be considered in Sec. IV.

IV. CALCULATION OF NONLINEAR SHEAR VISCOSITY

In this section we present approximate evaluations of (3.33) and (3.34). Since they involve complicated time-correlation functions, full microscopic evaluations of them are beyond our ability. We are here primarily concerned with contributions of long-wavelength fluctuations evaluated via the mode-coupling approximation.[17] For the shear dependence of $\eta(D)$ and for normal stress effects we shall see that indeed these long-wavelength fluctuations yield dominant contributions for the simple fluid under consideration. To simplify the calculations, we consider the fluid to be incompressible and consider only the fluctuations in transverse velocity, as these give the most important contributions to $\eta(D)-\eta(0)$ and to normal stress effects. We also suppose that the fluid is *not* near a phase transition. In the mode-coupling approximation, the averages are performed in the two steps: (i) Fix values of the gross variables (here the transverse velocity field) and average over all other microscopic degrees of freedom, and (ii) average over possible values of the gross variables. We now imagine that the first step was performed. Then $J^{\alpha\beta}(\vec{r})$ is replaced by its local equilibrium average with given transverse velocity field $\hat{\vec{v}}\equiv\vec{j}(\vec{r})/m$. In the two-mode-coupling approximation, we then find

$$J^{\alpha\beta}(\vec{r})-p\delta_{\alpha\beta}-mn_0\hat{v}^\alpha(\vec{r})\hat{v}^\beta(\vec{r}), \tag{4.1}$$

where the transverse nature of the local velocity field has been used to omit the term $(2T\alpha_s)^{-1}\times mn_0\hat{v}^2(\vec{r})\delta^{\alpha\beta}$ on the right-hand side of (4.1), where α_s is the adiabatic thermal expansion coefficient. This term, even if present, does not affect (3.33) and (3.34).

Let us now return to $\tilde{R}(t)$, (3.30), where J'^{xy} can be replaced by J^{xy} for the present incompressible fluid as pointed out at the end of Sec. III. As we shall show later, $(1-\mathcal{P}_0)$ in front of J'^{xy} can be replaced by 1 in the approximation used in this section. Here the problem is that $\mathcal{P}_0$ does not operate just on J'^{xy} but on J'^{xy} multiplied by everything that comes after. By (4.11) then $\tilde{R}(t)$ involves the time development of $\hat{v}^\alpha(\vec{r})$. The standard interpretation of the role of $\mathcal{P}_0$ in the time development operator $e^{it(1-\mathcal{P}_0)\tilde{L}}$ for the problems near equilibrium is that it removes any secular behavior that remains in the limit $t\to\infty$. We will assume that this is also true for the problems far from equilibrium, although this of course will need to be justified by future investigations. Then we may replace the effects of the time displacement upon $\hat{v}^\alpha$ by the hydrodynamic time evolution in the spirit of the mode-coupling theory in such a way that any secular behavior disappears. This can be done as follows: Take a Fourier component $\hat{v}^\alpha_{\vec{k}} = V^{-1/2}\int \hat{v}^\alpha(\vec{r})e^{-i\vec{k}\cdot\vec{r}}d\vec{r}$ rather than $\hat{v}^\alpha(\vec{r})$ and consider $\hat{v}^\alpha_{\vec{k}}(-t)=e^{-it\tilde{L}}\hat{v}^\alpha_{\vec{k}}e^{it\tilde{L}}$, $\alpha=x$ or y. The initial rate of change of $\hat{v}^\alpha_{\vec{k}}$ is

$$\left(\frac{d}{dt}\hat{v}^\alpha_{\vec{k}}(-t)\right)_{t=0} = -i[L,\hat{v}^\alpha_{\vec{k}}]-Di[L',\hat{v}^\alpha_{\vec{k}}]. \quad (4.2)$$

The first term on the right-hand side represents microscopic processes which after a certain time τ_0 which is long compared to the microscopic time scale of molecular collisions but short on a macroscopic scale give rise to viscous damping. Thus after the step (i) the first term will reduce to $-k^2(\eta/mn)\hat{v}^\alpha_{\vec{k}}$ where we note that in fact we are going backward in time, and we have dropped possible nonlinearities in the hydrodynamic equations. The second term of (4.2) can be directly evaluated to yield

$$-D\left(k_y\frac{\partial}{\partial k_x}\hat{v}^\alpha_{\vec{k}}-\delta_{\alpha y}\hat{v}^x_{\vec{k}}\right). \quad (4.3)$$

We then extrapolate this to $t>0$ and write

$$\frac{d}{dt}\hat{v}^\alpha_{\vec{k}}(-t) = -\frac{k^2\eta}{mn}\hat{v}^\alpha_{\vec{k}}(-t) - D\left(k_y\frac{\partial}{\partial k_x}\hat{v}^\alpha_{\vec{k}}(-t)-\delta_{\alpha y}\hat{v}^x_{\vec{k}}(-t)\right). \quad (4.4)$$

This, however, is not yet fully satisfactory, since the equation of motion (4.4) destroys the transverse character of $\hat{v}^\alpha_{\vec{k}}$ as one obtains from (4.4)

$$\frac{d}{dt}[\vec{k}\cdot\hat{\vec{v}}_{\vec{k}}(-t)] = 2k_y\hat{v}^x_{\vec{k}}(-t)$$

which does not vanish in general (where $\vec{k}\cdot\hat{\vec{v}}_k=0$ was used in evaluating the right-hand side). This can be remedied by adding to the right-hand side of (4.4) the term $-2Dk^{-2}k_yk_\alpha\hat{v}^x_{\vec{k}}(-t)$. This result can be also obtained by deriving from (4.4) the equation of motion for the explicitly transverse velocity $v^\alpha_{\vec{k}}-k^{-2}k_\alpha\vec{k}\cdot\vec{v}_{\vec{k}}$. Defining the dimensionless velocity $u^\alpha_{\vec{k}}$ by

$$u^\alpha_{\vec{k}} = (mn_0/k_BT)^{1/2}\hat{v}^\alpha_{\vec{k}}, \quad (4.5)$$

the equation of motion is finally obtained as

$$\frac{d}{dt}u^\alpha_{\vec{k}}(-t) = -k^2\frac{\eta}{mn_0}u^\alpha_{\vec{k}}(-t) - D\left(k_y\frac{\partial}{\partial k_x}u^\alpha_{\vec{k}}(-t) + \frac{2k_yk_\alpha}{k^2}u^x_{\vec{k}}(-t)-\delta_{\alpha y}u^x_{\vec{k}}(-t)\right). \quad (4.6)$$

Now, since by (4.1) we have after step (i)

$$I^{\alpha\beta}-Vp\delta_{\alpha\beta}\to mn_0\sum_{\vec{k}}\hat{v}^\alpha_{\vec{k}}\hat{v}^\beta_{-\vec{k}} = k_BT\sum_{\vec{k}}u^\alpha_{\vec{k}}u^\beta_{-\vec{k}} \quad (4.7)$$

and $\tilde{R}(t)$ contains only I^{xy}, we consider $u^x_{\vec{k}}u^y_{-\vec{k}}$, and then (4.4) gives

$$\frac{d}{dt}u^x_{\vec{k}}(-t)u^y_{-\vec{k}}(-t) = -2k^2\frac{\eta}{mn_0}u^x_{\vec{k}}(-t)u^y_{-\vec{k}}(-t) - D\left[\left(k_y\frac{\partial}{\partial k_x}+2\frac{k_xk_y}{k^2}\right)u^x_{\vec{k}}(-t)u^y_{-\vec{k}}(-t) - \left(1-2\frac{k_y^2}{k^2}\right)u^x_{\vec{k}}(-t)u^x_{-\vec{k}}(-t)\right]. \quad (4.8)$$

Likewise, we obtain

$$\frac{d}{dt}u^x_{\vec{k}}(-t)u^x_{-\vec{k}}(-t) = -2k^2\frac{\eta}{mn_0}u^x_{\vec{k}}(-t)u^x_{-\vec{k}}(-t) - D\left(k_y\frac{\partial}{\partial k_x}+4\frac{k_xk_y}{k^2}\right)\times u^x_{\vec{k}}(-t)u^x_{-\vec{k}}(-t). \quad (4.9)$$

Here and in what follows sums over $\vec{k}$ are limited by a certain cutoff which is much smaller than a microscopic wave number.

Introducing the two-component spinor $\psi(\vec{k}t)$ by

$$\psi(\vec{k},t) = \begin{pmatrix}\psi_1(\vec{k},t)\\ \psi_2(\vec{k},t)\end{pmatrix} \quad (4.10)$$

with

$$\psi_1(\vec{k},t) \equiv u^x_{\vec{k}}(-t)u^y_{-\vec{k}}(-t),$$
$$\psi_2(\vec{k}) \equiv u^x_{\vec{k}}(-t)u^x_{-\vec{k}}(-t), \tag{4.11}$$

(4.8) and (4.9) can be written as a single equation;

$$\frac{d}{dt}\psi(\vec{k}t) = -(\Gamma_{\vec{k}} + D\Lambda_{\vec{k}})\psi(\vec{k}t), \tag{4.12}$$

where

$$\Gamma_{\vec{k}} \equiv 2k^2\eta/mn_0, \tag{4.13}$$

$$\Lambda_{\vec{k}} \equiv k_y\frac{\partial}{\partial k_x} + \frac{k_x k_y}{k^2}(3-\sigma_z) + \left(\frac{k_y^2}{k^2} - \frac{1}{2}\right)(\sigma_x + i\sigma_y), \tag{4.14}$$

$\sigma_x, \sigma_y, \sigma_z$ being the Pauli spin matrices.

Now, the expressions (3.33) and (3.34) contain the time integral of $\tilde{R}(t)$ which in the present approximation can be obtained from the time integral of $\psi_1(\vec{k}t)$. Such a time integral exists only if the real parts of the eigenvalues of the operator $\Gamma_{\vec{k}} + D\Lambda_{\vec{k}}$ are positive definite. This problem is closely related to the problem of stability of laminar flow with the rate of shear D against small local velocity fluctuations whose temporal development is governed by (4.6). Introducing the two-component spinor $\phi(\vec{k}t)$ given by

$$\phi(\vec{k},t) \equiv \begin{pmatrix} u^y_{\vec{k}}(-t) \\ u^x_{\vec{k}}(-t) \end{pmatrix}, \tag{4.15}$$

(4.6) is written as

$$\frac{d}{dt}\phi(\vec{k},t) = -\Upsilon_{\vec{k}}\,\phi(\vec{k},t), \tag{4.16}$$

where

$$\Upsilon_{\vec{k}} = \tfrac{1}{2}\Gamma_{\vec{k}} + D(\Lambda_k - 2k_x k_y/k^2). \tag{4.17}$$

The ratio of the two terms in $\Upsilon_{\vec{k}}$ is roughly

$$\frac{D}{\frac{1}{2}\Gamma_k} = \frac{\partial v_y/\partial x}{k^2(\eta/mn)} \sim \frac{\bar{v}l}{\eta/mn}\frac{1}{(kl)^2} = \frac{R}{(kl)^2}, \tag{4.18}$$

where $\bar{v}$ is the average velocity and l is the macroscopic length over which the local velocity changes appreciably, and $R \equiv \bar{v}l/(\eta/mn)$ is the Reynolds number. If the laminar flow is stable, the real parts of all the eigenvalues of $\Upsilon_{\vec{k}}$ should be positive. If the instability is to occur, this will start at the velocity fluctuations with the smallest wave number where viscous damping is the smallest. The smallest wave number here is roughly equal to π/l. Since we are concerned with a stable laminar flow, we can conclude that all the fluctuations $u^x_{\vec{k}}$ and $u^y_{\vec{k}}$ decay in time. Hence $\psi(\vec{k},t)$ should also decay in time, that is, the real parts of all the eigenvalues of $\Gamma_{\vec{k}} + D\Lambda_{\vec{k}}$ should also be positive definite. This fact gives additional support for the validity of the replacement of the time displacement operator $e^{-i(1-\mathcal{P}_0)\mathcal{L}t}$ by the linearized hydrodynamic equations where no secular term remains.

We substitute these results into the expression of $\eta(D)$ which is now written as

$$\eta(D) = \frac{1}{D}\frac{mn_0}{V}\left\langle \sum_{\vec{k}} \hat{v}^x_{\vec{k}}\hat{v}^y_{-\vec{k}}\left[1 - \exp\left(-\beta mn_0 D\int_0^\infty \sum_{\vec{k}} \hat{v}^x_{\vec{k}}(-t)\hat{v}^y_{-\vec{k}}(-t)\,dt\right)\right]\right\rangle_{0c}, \tag{4.19}$$

where the cumulant average is taken regarding the pair $\hat{v}^x_{\vec{k}}\hat{v}^y_{-\vec{k}}$ as a single unit. We then find the following expression for $\eta(D)$ written in the matrix notation:

$$\eta(D) = \frac{1}{D}\frac{k_BT}{V}\left\langle \sum_{\vec{k}} \psi_1(\vec{k})\left[1 - \exp\left(-D\sum_{\vec{k}} \tilde{\nu}\cdot(\Gamma_{\vec{k}} + D\Lambda_{\vec{k}})^{-1}\cdot\psi(\vec{k})\right)\right]\right\rangle_{0c}, \tag{4.20}$$

where $\tilde{\nu}$ is a row matrix defined by

$$\tilde{\nu} \equiv (1,0). \tag{4.21}$$

In the same manner we also obtain for the normal stress effects,

$$P^{*xx}(D) - P^{*yy}(D) = \frac{k_BT}{V}\left\langle \sum_{\vec{k}}[\psi_2(\vec{k}) - \psi_3(\vec{k})]\left[\exp\left(-D\sum_{\vec{k}} \tilde{\nu}\cdot(\Gamma_{\vec{k}} + D\Lambda_{\vec{k}})^{-1}\cdot\psi(\vec{k})\right) - 1\right]\right\rangle_{0c}, \quad \text{etc.} \tag{4.22}$$

where

$$\psi_3(\vec{k}) \equiv u^y_{\vec{k}}u^y_{-\vec{k}}. \tag{4.23}$$

In our model incompressible fluid where density fluctuations are ignored, the probability distribution of fluctuation of $u^\alpha_{\vec{k}}$ takes the Gaussian form with

$$\langle u^\alpha_{\vec{k}}u^\beta_{\vec{k}'}\rangle_0 = \delta_{\vec{k},-\vec{k}'}(\delta_{\alpha\beta} - k_\alpha k_\beta/k^2). \tag{4.24}$$

This fact can be used profitably to evaluate (4.20) and (4.22) where each term in the expansions of (4.20) and (4.22) in powers of $\beta mn_0 D$ contains factors like

$$\langle \delta(\hat{v}^{\alpha}_{\vec{k}} \hat{v}^{\beta}_{-\vec{k}}) \delta(\hat{v}^{\gamma}_{\vec{k}} \hat{v}^{\delta}_{-\vec{k}}) \cdots \delta(\hat{v}^{\sigma}_{\vec{k}} \hat{v}^{\tau}_{-\vec{k}}) \rangle_{0c} = \sum \langle \hat{v}^{\beta}_{-\vec{k}} \hat{v}^{\gamma}_{\vec{k}} \rangle_0 \cdots \langle \hat{v}^{\tau}_{-\vec{k}} \hat{v}^{\alpha}_{\vec{k}} \rangle_0 . \quad (4.25)$$

Here $\delta X = X - \langle X \rangle_0$ and the sum on the right-hand side is over all the ways in which products of the $\hat{v}$'s are factorized into pairs in such a way that all the $\delta(\hat{v}\hat{v})$'s on the left-hand sides are connected.

We now discuss the effects of $\mathcal{P}_0$ in front of J'^{xy} in (3.30) which we have temporarily ignored. $\mathcal{P}_0$ will then produce a term like

$$\langle \delta(\hat{v}^{\alpha}_{\vec{k}} \hat{v}^{\beta}_{-\vec{k}}) \delta(\hat{v}^{\gamma}_{\vec{k}} \hat{v}^{\delta}_{-\vec{k}}) \cdots \delta(\hat{v}^{\epsilon}_{\vec{k}} \hat{v}^{\xi}_{-\vec{k}}) \mathcal{P}_0 \delta(\hat{v}^{\mu}_{\vec{k}'} \hat{v}^{\nu}_{-\vec{k}'}) \cdots \delta(\hat{v}^{\sigma}_{\vec{k}'} \hat{v}^{\tau}_{-\vec{k}'}) \rangle_{0c}$$
$$= \sum_{\lambda} \langle \delta(\hat{v}^{\alpha}_{\vec{k}} \hat{v}^{\beta}_{-\vec{k}}) \delta(\hat{v}^{\gamma}_{\vec{k}} \hat{v}^{\delta}_{-\vec{k}}) \cdots \delta(\hat{v}^{\epsilon}_{\vec{k}} \hat{v}^{\xi}_{-\vec{k}}) \delta A_0^{\lambda *} \rangle_{0c} \langle |\delta A_0^{\lambda}|^2 \rangle^{-1} \langle \delta A_0^{\lambda} \delta(\hat{v}^{\mu}_{\vec{k}'} \hat{v}^{\nu}_{-\vec{k}'}) \cdots \delta(\hat{v}^{\sigma}_{\vec{k}'} \hat{v}^{\tau}_{-\vec{k}'}) \rangle_{0c} , \quad (4.26)$$

where the A_0^{λ}'s are the spatial integrals of the densities of gross variables. Then, we have

$$\langle \delta A_0^{\lambda} \delta(\hat{v}^{\mu}_{\vec{k}} \hat{v}^{\nu}_{-\vec{k}}) \cdots \rangle_{0c} = \frac{\partial}{\partial b^{\lambda}} \langle \delta(\hat{v}^{\mu}_{\vec{k}} \hat{v}^{\nu}_{-\vec{k}}) \cdots \rangle_{0c} \quad (4.27)$$

and

$$\langle |\delta A_0^{\lambda}|^2 \rangle \sim V , \quad (4.28)$$

where b^{λ} is the field conjugate to A_0^{λ}. Comparing these results with (4.25), we note that a projection operator $\mathcal{P}_0$ in front of J'^{xy} of (3.30) produces terms having an extra factor V^{-1}. However, the sum over $\vec{k}'$ produces another factor V. Hence $\mathcal{P}_0$ produces additional finite contributions. On the other hand, as we shall soon see, the terms we have retained earlier involve a single summation over $\vec{k}$, and give dominant divergent contributions when one tries to expand the final results in powers of D. In comparison the new terms produced by $\mathcal{P}_0$ give only finite or less divergent contributions and hence we shall drop these terms, since their magnitude will be smaller by the factor of the order $k_0^3/n_0 \sim (Dmn_0^{1/3}/\eta)^{3/2} \ll 1$.

The actual evaluation of (4.20) and (4.22) requires rather lengthy algebra and is carried out in Appendix B; the final numerical results are summarized here:

$$\Delta\eta(D) \equiv \eta(D) - \eta(0) \cong -5.1 \times 10^{-3} k_B T (mn_0/\eta)^{3/2} D^{1/2} , \quad (4.29)$$

$$P^{xx}(D) - P^{yy}(D) \cong -6.7 \times 10^{-3} k_B T (mn_0 D/\eta)^{3/2} , \quad (4.30)$$

$$P^{xx}(D) - P^{zz}(D) \cong -2.9 \times 10^{-3} k_B T (mn_0 D/\eta)^{3/2} . \quad (4.31)$$

The fractional powers of D appearing in these results can be understood also as follows: First, let us formally expand (4.20) and (4.22) in powers of D. Invariance under rotation by 180° about the x axis and the simultaneous change of sign of D tells us that $\Delta\eta(D)$ as well as $P^{ii}(D)$ are even functions of D. Thus the series contains only the terms with even powers in D. Then a term in the expansion of (4.29) takes the following general form:

$$\frac{k_B T}{V} \sum_{\vec{k}} \langle \psi_1(\vec{k}) \vec{v} \cdot \Gamma^{-1}_{\vec{k}} D \lambda_{\alpha_1} \Gamma^{-1}_{\vec{k}} \times D \lambda_{\alpha_2} \Gamma^{-1}_{\vec{k}} \cdots D \lambda_{\alpha_{2n}} \Gamma^{-1}_{\vec{k}} \psi(\vec{k}) \rangle_{0c} , \quad (4.32)$$

where the matrices λ_α with $\alpha = 1$ or 2 are given by

$$\lambda_1 = \Lambda_{\vec{k}} , \quad \lambda_2 = \begin{pmatrix} \psi_1(\vec{k}) & 0 \\ \psi_2(\vec{k}) & 0 \end{pmatrix} . \quad (4.33)$$

Equation (4.32) can be estimated by noting that ψ_1, ψ_2, $\vec{v}$, and λ_α are quantities of order unity. Hence (4.32) has the magnitude

$$[k_B T D^2 m^3 n_0^3 / k_0^3 \eta^3][Dmn_0/k_0^2 \eta]^{2(n-1)} , \quad n = 1, 2, 3, \ldots , \quad (4.34)$$

where k_0 is the small wave number cutoff in the summation over $\vec{k}$ which is necessary to prevent the divergences in each term of the expansion at small $\vec{k}$. These divergences arise because the transverse velocity fluctuation with wave number $\vec{k}$ is allowed to decay with the decay rate $\frac{1}{2}\Gamma_{\vec{k}}$. On the other hand, the discussion made in connection with (4.18) indicates that the velocity gradient D starts to interfere with the decay of the transverse velocity at the wave number where the ratio (4.18) becomes of the order unity. Precisely the same ratio with $k = k_0$ now appears as the expansion parameter in our series. Hence the appropriate cutoff wave number is

$$k_0 = (\text{const})(Dmn_0/\eta)^{1/2} . \quad (4.35)$$

Then each term in the expansion has the order of magnitude value $k_B T (mn_0/\eta)^{3/2} D^{1/2}$ which is precisely what is found in Appendix B. Practically the same arguments go through for $P^{xx} - P^{yy}$ and $P^{xx} - P^{zz}$ as well.

In Ref. 6 as well as in Ref. 18 the nonlinear shear viscosity in the critical mixture was studied where only the mode coupling to the concentration fluctuations was taken into account, on the ground that the concentration fluctuations exhibit critical enhancement. The subsequent study has shown that the contributions from concentration fluctuations are much smaller than those considered in these works.[19] The critical anomalies considered in these works now appear to be practically masked by the transverse velocity fluctuations considered in the present work.

V. CONCLUDING REMARKS

In the preceding sections we first derived formally exact equations of motion for gross variables valid arbitrarily far from equilibrium which are extensions of the time-correlation approach in transport theory. On the other hand, in the case of a gas one can study the same problems on the basis of the Boltzmann equation and its extensions to higher densities. This allows us to compare these two approaches in transport theory on a larger scale than has been done in the past, where such comparisons have been limited to linear transport phenomena.[1,20] For such a comparison one needs to develop a systematic density expansion technique for the general expression for the average steady-state flux (2.39) which is similar to the one employed for the linear transport coefficients.[20]

We have applied the general formalism to nonlinear shear viscosity and normal stress effects in fluids where the long-time tails in time-correlation functions produce new nonanalytic terms in the rate of shear. Simple numerical estimates show that unfortunately these nonanalytic terms are too small to be directly observable under ordinary experimental conditions, but may have some observable effects under extreme conditions such as correspond to shock waves.

The mode-coupling approximation used here does not assume a low-density situation. On the other hand, the question of the long-time tails in the low-density gas is an interesting one.[21,22] The same kind of long-time-tail effects which were taken into account in the present work have been recovered in the context of ordinary time correlation functions for low-density gases by considering the so-called ring processes.[23] On the other hand the same long-time effects can be derived by regarding the nonlinear Boltzmann equation as a sort of kinetic equation of the mode coupling theory.[17,22] However, the contribution of these long-time tails to transport coefficients are of higher orders in the density than those predicted by the Chapman-Enskog theory. Clearly, the analogous, more elaborate consideration is needed to compare our nonlinear transport theory with the predictions of the Boltzmann equation and its extensions to higher densities.

Also, it would be of considerable interest to study the cases where at least two kinds of thermodynamic driving forces like a velocity gradient and a temperature gradient are present, to see how the interference of different transport processes are affected by the long-time-tail contributions.

Recently the problem of nonanalytic corrections to the linearized hydrodynamic equations has received a great deal of attention, in an attempt to study the range of validity of classical hydrodynamics. In this case transport coefficients exhibit nonanalytic dependences upon the frequency and the wave number of an external disturbance.[21] Such attempts are incomplete as long as nonlinear effects are omitted. The present paper demonstrates that there is also a nonanalyticity associated with the amplitude of an external disturbance, and the method developed here provides a way to investigate this important problem.

Finally, we would like to note that the general formalism presented in Sec. II also applies for the phase-space distribution function itself. In particular one can derive the following identity for $\rho(t)$ which corresponds to (2.36):

$$\begin{aligned}\rho(t)=\rho_l(t)\bigg\{&\exp_+\left[-\int_{t_0}^t R(ts)ds\right]\\&+\int_{t_0}^t ds\,\exp_+\left[-\int_s^t R(ts')ds'\right]\\&\times U(ts)\,\delta_s\vec{A}\cdot\dot{\vec{b}}(s)\bigg\}\cdot 1\\&+U_l(t,t_0)[1-\mathcal{P}_l(t_0)]\rho(t_0),\end{aligned}\qquad(5.1)$$

where t_0 is an arbitrary initial time which we can take to be $-\infty$. For a steady state this simplifies to

$$\rho=\rho_l\exp_-\left[-\int_0^\infty R(t)dt\right]\cdot 1.\qquad(5.2)$$

Using this one can study not only the average behavior but also the fluctuations which occur in a steady state or a slowly varying state of the system. We hope to discuss these and related problems in the near future.

APPENDIX A

The property (2.6a) is demonstrated for an arbitrary phase-space function as follows:

$$\mathcal{P}_l(t)\mathcal{P}_l(t')X=\mathcal{P}_l(t)\left[\left(\rho_l(t')-\frac{\partial\rho_l(t')}{\partial\langle\vec{A}\rangle_{t'}}\cdot\langle\vec{A}\rangle_{t'}\right)\,\mathrm{Tr}X+\frac{\partial\rho_l(t')}{\partial\langle\vec{A}\rangle_{t'}}\cdot\mathrm{Tr}AX\right]$$
$$=\left(\rho_l(t)-\frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\langle\vec{A}\rangle_t\right)\mathrm{Tr}X+\frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\left[\left(\langle\vec{A}\rangle_{t'}-\frac{\partial\langle\vec{A}\rangle_{t'}}{\partial\langle\vec{A}\rangle_{t'}}\cdot\langle\vec{A}\rangle_{t'}\right)\mathrm{Tr}X+\frac{\partial\langle\vec{A}\rangle_{t'}}{\partial\langle\vec{A}\rangle_{t'}}\cdot\mathrm{Tr}\vec{A}X\right]$$
$$=\left(\rho_l(t)-\frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\langle\vec{A}\rangle_t\right)\mathrm{Tr}X+\frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\mathrm{Tr}\vec{A}X=\mathcal{P}_l(t)X. \tag{A1}$$

In order to demonstrate (2.6c) we differentiate (2.6b) with respect to t. Then we only have to show that $\dot{\mathcal{P}}_l(t)\rho(t)$ vanishes, which follows:

$$\dot{\mathcal{P}}_l(t)\rho(t)=\left[\dot\rho_l(t)-\frac{\partial}{\partial t}\left(\frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\cdot\langle\vec{A}\rangle_t\right)\right]+\left(\frac{\partial}{\partial t}\,\frac{\partial\rho_l(t)}{\partial\langle\vec{A}\rangle_t}\right)\cdot\langle\vec{A}\rangle_t=\dot\rho_l(t)-\frac{\partial\rho_l(t)}{\partial\langle A\rangle_t}\cdot\frac{\partial}{\partial t}\langle\vec{A}\rangle_t=0. \tag{A2}$$

The last step is the consequence of the fact that $\rho_l(t)$ depends upon time only through $\langle A\rangle_t$.

APPENDIX B

Here we shall evaluate (4.20) and (4.22). First introduce a two-component function $\xi(\vec{k})$ of $\vec{k}$ by

$$\xi(\vec{k})=\begin{pmatrix}\xi_1(\vec{k})\\ \xi_2(\vec{k})\end{pmatrix} \tag{B1}$$

which satisfies the following equation:

$$(\Gamma_{\vec{k}}+D\Lambda_{\vec{k}})\cdot\xi(\vec{k})=\psi(\vec{k}), \tag{B2}$$

or explicitly,

$$\left(\frac{\partial}{\partial k_x}+2\frac{k_x}{k^2}+2g\frac{k^2}{k_y}\right)\xi_1(\vec{k})+\left(2\frac{k_y}{k^2}-\frac{1}{k_y}\right)\xi_2(\vec{k})=\frac{1}{Dk_y}\psi_1(\vec{k}), \tag{B3}$$

$$\left(\frac{\partial}{\partial k_x}+4\frac{k_x}{k^2}+2g\frac{k^2}{k_y}\right)\xi_2(\vec{k})=\frac{1}{Dk_y}\psi_2(\vec{k}), \tag{B4}$$

where

$$g\equiv\eta/(mn_0D).$$

Then the exponentials in (4.20) and (4.22) become simply

$$\exp\left(-D\sum_{\vec{k}}\xi_1(\vec{k})\right). \tag{B5}$$

Equations (B3) and (B4) are solved to yield

$$\xi_2(\vec{k})=\int_{-\infty\times Dk_y}^{k_x}dk_x'\left(\frac{k'}{k}\right)^4\exp\left(\frac{2g}{k_y}\left[k_x'(k'^2-\tfrac{2}{3}k_x'^2)-k_x(k^2-\tfrac{2}{3}k_x^2)\right]\right)\frac{\psi_2(\vec{k}')}{Dk_y}$$
$$+\lim_{k_{0x}\to-\infty\times Dk_y}\left[\left(\frac{k_0}{k}\right)^4\exp\left(\frac{2g}{k_y}k_{0x}(k_0^2-\tfrac{2}{3}k_{0x}^2)-\frac{2g}{k_y}k_x(k^2-\tfrac{2}{3}k_x^2)\xi_2(\vec{k}_0)\right)\right], \tag{B6}$$

where $k_{0y}=k_y'=k_y$ and $k_{0z}=k_z'=k_z$, and

$$\xi_1(\vec{k})=\int_{-\infty\times Dk_y}^{k_x}dk_x'\left(\frac{k'}{k}\right)^2\exp\left(\frac{2g}{k_y}\left[k_x'(k'^2-\tfrac{2}{3}k_x'^2)-k_x(k^2-\tfrac{2}{3}k_x^2)\right]\right)$$
$$\times\left[\frac{\psi_1(\vec{k}')}{Dk_y}+\left(\frac{1}{k_y}-\frac{2k_y}{k'^2}\right)\xi_2(\vec{k}')\right]+\lim_{k_{0x}\to-\infty\times Dk_y}\left[\left(\frac{k_0}{k}\right)^2\exp\left(\frac{2g}{k_y}k_{0x}(k_0^2-\tfrac{2}{3}k_{0x}^2)-\frac{2g}{k_y}k_x(k^2-\tfrac{2}{3}k_x^2)\right)\xi_1(\vec{k}_0)\right]. \tag{B7}$$

The lower limits of integrations in (B6) and (B7) are so chosen to make the integrals convergent. We observe that unless $\xi_2'(\vec{k}_0)$ increases exponentially as $k_{0x}\to-\infty\times Dk_y$, the second terms of (B6) and (B7) vanish, which we assume to be the case here. We can then write the exponential (B5), after some algebra, as

$$\exp\left(-\sum_{\vec{k}}F(\vec{k})u_{\vec{k}}^x u_{-\vec{k}}^y-\sum_{\vec{k}}F_x(\vec{k})u_{\vec{k}}^x u_{-\vec{k}}^x\right), \tag{B8}$$

where

$$F(\vec{k})\equiv\frac{1}{k_y}\int_{k_x}^{\infty\times Dk_y}dk_x'\left(\frac{k}{k'}\right)^2\exp\left(\frac{2g}{k_y}\left[k_x(k^2-\tfrac{2}{3}k_x^2)-k_x'(k'^2-\tfrac{2}{3}k_x'^2)\right]\right) \tag{B9}$$

and

$$F_x(\vec{k}) \equiv \int_{k_x}^{\infty \times Dk_y} dk'_x \frac{k^4}{k'^2}\left[\frac{(k_z/k_y)^2}{(k_y^2+k_z^2)^{3/2}}\tan^{-1}\left(\frac{(k'_x-k_x)(k_y^2+k_z^2)^{1/2}}{k_y^2+k_z^2+k_x k'_x}\right) - \frac{k'_x-k_x}{(kk')^2}\left(1-\frac{k_x k'_x}{k_y^2+k_z^2}\right)\right]\exp\left(\frac{2g}{k_y}\left[k_x(k^2-\tfrac{2}{3}k_x^2)-k'_x(k'^2-\tfrac{2}{3}k_x'^2)\right]\right). \quad \text{(B10)}$$

Equation (4.20) is now written as

$$\eta(D) = \frac{k_B T}{DV}\left\langle \sum_{\vec{k}} u^x_{\vec{k}} u^y_{-\vec{k}}\left[1-\exp\left(-\sum_{\vec{k}} F(\vec{k}) u^x_{\vec{k}} u^y_{-\vec{k}} - \sum_{\vec{k}} F_x(\vec{k}) u^x_{\vec{k}} u^x_{-\vec{k}}\right)\right]\right\rangle_{0c}$$

$$= \frac{k_B T}{2DV}\sum_{\vec{k}} \frac{\partial}{\partial F(\vec{k})}\left\langle \exp\left(-\sum_{\vec{k}} F(\vec{k}) u^x_{\vec{k}} u^y_{-\vec{k}} - \sum_{\vec{k}} F_x(\vec{k}) u^x_{\vec{k}} u^x_{-\vec{k}}\right)\right\rangle_{0c}$$

$$= \frac{k_B T}{2DV}\sum_{\vec{k}} \frac{\partial}{\partial F(\vec{k})} \ln\left\langle \exp\left(-\sum_{\vec{k}} F(\vec{k}) u^x_{\vec{k}} u^y_{-\vec{k}} - \sum_{\vec{k}} F_x(\vec{k}) u^x_{\vec{k}} u^x_{-\vec{k}}\right)\right\rangle_{0}, \quad \text{(B11)}$$

where we have made use of the fact that $F(\vec{k})$ is an even function of $\vec{k}$ and have also used a general relationship

$$\langle e^X\rangle_c = 1 + \ln\langle e^X\rangle. \quad \text{(B12)}$$

Here $\langle e^X\rangle_c$ is the connected average where each factor X in the power-series expansion of e^X is correlated. Similarly we find

$$P^{xx}(D) - P^{yy}(D) = \Delta^x(D) - \Delta^y(D), \quad P^{xx}(D) - P^{zz}(D) = \Delta^x(D) - \Delta^z(D), \quad \text{(B13)}$$

where

$$\Delta^\alpha(D) \equiv \frac{k_B T}{V}\sum_{\vec{k}}\left\langle u^\alpha_{\vec{k}} u^\alpha_{-\vec{k}}\left[\exp\left(-\sum_{\vec{k}} F(\vec{k}) u^x_{\vec{k}} u^y_{-\vec{k}} - \sum_{\vec{k}} F_x(\vec{k}) u^x_{\vec{k}} u^x_{-\vec{k}}\right) - 1\right]\right\rangle_{0c}$$

$$= -\frac{k_B T}{V}\sum_{\vec{k}}\left[1-\left(\frac{k_\alpha}{k}\right)^2 + \frac{1}{2}\frac{\partial}{\partial F_\alpha(\vec{k})}\ln\left\langle \exp\left(-\sum_{\vec{k}} F(\vec{k}) u^x_{\vec{k}} u^y_{-\vec{k}} - \sum_{\beta}^{xyz}\sum_{\vec{k}} F_\beta(\vec{k}) u^\beta_{\vec{k}} u^\beta_{-\vec{k}}\right)\right\rangle_0\right] \quad (\alpha = x, y, z) \quad \text{(B14)}$$

evaluated at $F_y(\vec{k}) = F_z(\vec{k}) = 0$. Here we have made use of the fact that $F_x(\vec{k})$ is also an even function of $\vec{k}$, and have supposed that $F_y(\vec{k})$ and $F_z(\vec{k})$ are some even functions of $\vec{k}$ to be set equal to zero afterwards.

The averages are taken with respect to the equilibrium distribution function for transverse velocities, $P_0(\{\vec{u}\})$, which we slightly modify by including small longitudinal components of velocity as follows:

$$P_0(\{u\}) = \mathfrak{N}\exp\left(-\frac{1}{2}\sum_{\vec{k}}\sum_{\alpha\beta} u^\alpha_{\vec{k}}\,[t_\epsilon^{-1}(\vec{k})]^{\alpha\beta} u^\beta_{-\vec{k}}\right), \quad \text{(B15)}$$

where $t_\epsilon(\vec{k})$ is the nonsingular 3×3 matrix defined by

$$[t_\epsilon(\vec{k})]^{\alpha\beta} = \delta_{\alpha\beta} - (1-\epsilon^2)\hat{k}_\alpha \hat{k}_\beta \quad \text{(B16)}$$

with $\hat{k}_\alpha \equiv k_\alpha/k$, $\mathfrak{N}$ the normalization factor, and ϵ a small positive number. We then find

$$\langle u^\alpha_{\vec{k}} u^\beta_{-\vec{k}'}\rangle_0 = \delta_{\vec{k},\vec{k}'}[t_\epsilon(\vec{k})]^{\alpha\beta} \quad \text{(B17)}$$

which reduces to (4.20) as $\epsilon \to 0^+$. The inverse of $t_\epsilon(\vec{k})$ is readily obtained as

$$[t_\epsilon^{-1}(\vec{k})]^{\alpha\beta} = \delta_{\alpha\beta} + (\epsilon^{-2}-1)\hat{k}_\alpha\hat{k}_\beta. \quad \text{(B18)}$$

The equilibrium average appearing in (B14) now becomes

$$\mathfrak{N}\prod_{\vec{k},\gamma}\int du^\gamma_{\vec{k}}\exp\left(-\frac{1}{2}\sum_{\vec{k}}\sum_{\alpha\beta} M^{\alpha\beta}(\vec{k}) u^\alpha_{\vec{k}} u^\beta_{-\vec{k}}\right), \quad \text{(B19)}$$

where

$$M^{\alpha\beta}(\vec{k}) \equiv \delta_{\alpha\beta} + (\epsilon^{-2}-1)\hat{k}_\alpha\hat{k}_\beta + F(\vec{k})(\delta_{\alpha x}\delta_{\beta y} + \delta_{\alpha y}\delta_{\beta x}) + 2\delta_{\alpha\beta}F_\alpha(\vec{k}). \quad \text{(B20)}$$

We may now interpret (B19) as the probability distribution function of velocity apart from the normalization in the presence of a constant rate of shear. The stability of such a laminar flow requires the probability distribution to be maximum at $\vec{u}_{\vec{k}} = 0$ $(\vec{k} \neq 0)$, which in turn requires the matrix $M^{\alpha\beta}(\vec{k})$ to be positive definite, which seems to be supported by the later numerical study.

Therefore, we obtain

$$\ln\left\langle \exp\left(-\sum_{\vec{k}} F(\vec{k}) u^x_{\vec{k}} u^y_{-\vec{k}} - \sum_{\alpha}^{xyz}\sum_{\vec{k}} F_\alpha(\vec{k}) u^\alpha_{\vec{k}} u^\alpha_{-\vec{k}}\right)\right\rangle_0 = (\text{const}) - \frac{1}{2}\sum_{\vec{k}} \ln\det M(\vec{k}), \quad \text{(B21)}$$

where const is a constant independent of $F(\vec{k})$ and $F_\alpha(\vec{k})$ and thus does not contribute to $\Delta^\alpha(D)$ and $\eta(D)$. For small ϵ, the leading term of $M(\vec{k})$ is of the order of ϵ^{-2}, that is,

$$\det M(\vec{k}) = \epsilon^{-2}\{1 + 2F_x(1-\hat{k}_x^2) + 2F_y(1-\hat{k}_y^2) + 2F_z(1-\hat{k}_z^2) - 2F\hat{k}_x\hat{k}_y - F^2\hat{k}_z^2 + 4F_xF_y\hat{k}_z^2 + 4F_yF_z\hat{k}_x^2 + 4F_zF_x\hat{k}_y^2 - 4FF_z\hat{k}_x\hat{k}_y\} + \cdots, \quad \text{(B22)}$$

where $\cdots$ stands for higher-order terms in ϵ which need not be considered. (B21) and (B22) are substituted into (B11) and (B14) to yield the following results:

$$\eta(D) = \frac{k_BT}{D}\frac{1}{(2\pi)^3}\int d\vec{k}\,\frac{\hat{k}_x\hat{k}_y + F(\vec{k})\hat{k}_z^2}{\mathfrak{D}(\vec{k})}, \quad \text{(B23)}$$

$$\Delta^x(D) = \frac{k_BT}{(2\pi)^3}\int d\vec{k}\,(1-\hat{k}_x^2)\left(\frac{1}{\mathfrak{D}(\vec{k})} - 1\right), \quad \text{(B24)}$$

$$\Delta^y(D) = \frac{k_BT}{(2\pi)^3}\int d\vec{k}\left(\frac{1-\hat{k}_y^2 + 2F_x\hat{k}_z^2}{\mathfrak{D}(\vec{k})} - 1 + \hat{k}_y^2\right), \quad \text{(B25)}$$

$$\Delta^z(D) = \frac{k_BT}{(2\pi)^3}\int d\vec{k}\left(\frac{1-\hat{k}_z^2 + 2F_x\hat{k}_y^2 - 2F\hat{k}_x\hat{k}_y}{\mathfrak{D}(\vec{k})} - 1 + \hat{k}_z^2\right), \quad \text{(B26)}$$

where

$$\mathfrak{D}(\vec{k}) \equiv 1 + 2F_x(\vec{k})(1-\hat{k}_x^2) - F^2(\vec{k})\hat{k}_z^2 - 2F(\vec{k})\hat{k}_x\hat{k}_y. \quad \text{(B27)}$$

In order to find out how $\eta(D)$ and $\Delta^\alpha(D)$ behave we first need to know the properties of the functions $F(\vec{k})$ and $F_x(\vec{k})$. First we note that

$$F(\vec{k}, -D) = -F(k_x, -k_y, k_z, D), \quad \text{(B28)}$$

$$F_x(\vec{k}, -D) = F_x(k_x, -k_y, k_z, D), \quad \text{(B29)}$$

which implies that $\eta(D)$ and $\Delta^\alpha(D)$ are the even functions of D as are expected. Hence without loss of generality we can assume D to be positive. We then introduce the dimensionless wave vector $\vec{l}$ by

$$\vec{l} = g^{1/2}\vec{k} \quad \text{(B30)}$$

and change the variable of integration from k_x' to s given by

$$k_x' = k_x + k_y s \quad \text{(B31)}$$

to obtain

$$F(\vec{k}) = \hat{F}(\vec{l}) \equiv \int_0^\infty ds\,(1 + 2\hat{l}_x\hat{l}_y s + \hat{l}_y^2 s^2)^{-1}\exp[-2l^2 s(1 + \hat{l}_x\hat{l}_y s + \tfrac{1}{3}\hat{l}_y^2 s^2)], \quad \text{(B32)}$$

$$F_x(\vec{k}) = \hat{F}_x(\vec{l}) \equiv \int_0^\infty ds\,\frac{\hat{l}_y}{1 + 2\hat{l}_x\hat{l}_y s + \hat{l}_y^2 s^2}\left[\frac{(\hat{l}_z/\hat{l}_y)^2}{(1-\hat{l}_x^2)^{3/2}}\tan^{-1}\frac{\hat{l}_y(1-\hat{l}_x^2)^{1/2}}{1 + \hat{l}_x\hat{l}_y s} - \frac{\hat{l}_y s}{1 + 2\hat{l}_x\hat{l}_y s + \hat{l}_y^2 s^2}\left(1 - \frac{(\hat{l}_x + \hat{l}_y s)\hat{l}_x}{1-\hat{l}_x^2}\right)\right]\exp\{-2l^2 s[1 + \hat{l}_x\hat{l}_y s + \tfrac{1}{3}\hat{l}_y^2 s^2]\}, \quad \text{(B33)}$$

where $\hat{l}$ is the unit vector. It is useful to investigate the asymptotic behavior of these functions for both $l \gg 1$ and $l \ll 1$. For $l \gg 1$ the integrals over s are limited to small s less than $O(1/l^2)$ and we find

$$\hat{F}(\vec{l}) \cong \tfrac{1}{2}l^2 \qquad (l \gg 1 \text{ or } l^2 \gg |\hat{l}_y|), \quad \text{(B34)}$$

$$\hat{F}_x(\vec{l}) \cong \frac{\hat{l}_y}{1-2\hat{l}_x^2}\left[\left(\frac{\hat{l}_z}{\hat{l}_y}\right)^2 - 1 + 2\hat{l}_x^2\right]\frac{1}{(2l^2)^2} \qquad (l \gg 1 \text{ or } l^2 \gg |\hat{l}_y|), \quad \text{(B35)}$$

where we have noted that these asymptotic expressions also hold even if l is not large, provided that $|\hat{l}_y|$ is small enough so that the quantity $|\hat{l}_y|s$ can be ignored.

Turning now to the region $l \ll 1$ we only consider the case where $l^2 \ll |\hat{l}_y|$ since another extreme case $l^2 \gg |\hat{l}_y|$ was already considered. Then, the cutoff in the integrals over s is provided by the denominator $1 + 2\hat{l}_x\hat{l}_y s + \hat{l}_y^2 s^2$ which limits s to be less than $O(|\hat{l}_y|^{-1})$, and the exponential factors in the integrands reduce to unity. We thus obtain

$$\hat{F}(\vec{l}) \cong \frac{1}{\hat{l}_y(1-\hat{l}_x^2)^{1/2}}\left(\frac{\hat{l}_y}{|\hat{l}_y|}\frac{\pi}{2} - \tan^{-1}\frac{\hat{l}_x}{(1-\hat{l}_x^2)^{1/2}}\right) \quad (l \ll 1 \text{ and } |\hat{l}_y| \gg l^2), \quad \text{(B36)}$$

$$\hat{F}_x(\vec{l}) \equiv \mathfrak{F}_x(\hat{\vec{l}}) \equiv \int_0^\infty \frac{\hat{l}_y}{1+2\hat{l}_x\hat{l}_y s+\hat{l}_y^2 s^2}\left[\frac{(\hat{l}_z/\hat{l}_y)^2}{(1-\hat{l}_x^2)^{3/2}}\tan^{-1}\frac{\hat{l}_y(1-\hat{l}_x^2)^{1/2}}{1+\hat{l}_x\hat{l}_y s}\,s - \frac{\hat{l}_y s}{1+2\hat{l}_x\hat{l}_y s+\hat{l}_y^2 s^2}\left(1-\frac{(\hat{l}_x+\hat{l}_y s)}{1-\hat{l}_x^2}\right)\right] \quad (l \ll 1 \text{ and } |\hat{l}_y| \gg l^2). \tag{B37}$$

From these asymptotic behavior we find that the integral for $\eta(D)$ given by (B23) contains a divergence at large values of k. The source of this is easy to identify. Namely, the mode-coupling contributions coming from short-wavelength fluctuations dominate $\eta(D)$, which should be also true for the linear shear viscosity $\eta(0)$. Therefore, from now on we shift our attention to the difference

$$\Delta\eta(D) \equiv \eta(D)-\eta(0) = \frac{k_B T}{(2\pi)^3} D^{1/2}\left(\frac{\rho}{\eta}\right)^{3/2}\int d\vec{l}\left(\frac{\hat{l}_z^2\hat{F}(\vec{l})+\hat{l}_x\hat{l}_y}{\hat{\mathfrak{D}}(\vec{l})} - \hat{l}_x\hat{l}_y - \frac{\hat{l}_z^2}{2l^2} - \frac{(\hat{l}_x\hat{l}_y)^2}{l^2}\right), \tag{B38}$$

where $\rho = mn$ and

$$\hat{\mathfrak{D}}(\vec{l}) \equiv 1+2\hat{F}_x(\vec{l})(1-\hat{l}_x^2) - \hat{F}^2(\vec{l})\hat{l}_z^2 - 2\hat{F}(\vec{l})\hat{l}_x\hat{l}_y, \tag{B39}$$

and certain vanishing terms are subtracted from (B38) to assure the convergence of the integral. In this form the integral is manifestly convergent and reduces to a finite number independent of all the variables D, n, etc. It is noteworthy that $\Delta\eta(D)$ does not have a power-series expansion in D^2 as one might naively expect from symmetry. The reason for this is again not difficult to see. If we formally expand $\eta(D)$ given by, say, (4.20) and examine the coefficients, the integrals over k which enter them are shown to diverge at small wave numbers indicating the dominance of long-wavelength fluctuations as was discussed in Sec. IV. This is another manifestation of such long-wavelength fluctuations which contributed to the long-time tails of time-correlation functions.[6,8]

In the similar manner we can reduce the expressions for $\Delta^\alpha(D)$ to the manifestly convergent forms as follows:

$$\Delta^x(D) = \frac{k_B T}{(2\pi)^3}\left(\frac{\rho D}{\eta}\right)^{3/2}\int d\vec{l}\,(1-\hat{l}_x^2)\left(\frac{1}{\hat{\mathfrak{D}}(\vec{l})} - 1 - \frac{\hat{l}_x\hat{l}_y}{l^2}\right), \tag{B40}$$

$$\Delta^y(D) = \frac{k_B T}{(2\pi)^3}\left(\frac{\rho D}{\eta}\right)^{3/2}\int d\vec{l}\left[(1-\hat{l}_y^2)\left(\frac{1}{\hat{\mathfrak{D}}(\vec{l})} - 1 - \frac{\hat{l}_x\hat{l}_y}{l^2}\right) + \frac{2\hat{F}_x(\vec{l})\hat{l}_z^2}{\hat{\mathfrak{D}}(\vec{l})}\right], \tag{B41}$$

$$\Delta^z(D) = \frac{k_B T}{(2\pi)^3}\left(\frac{\rho D}{\eta}\right)^{3/2}\int d\vec{l}\left[(1-\hat{l}_z^2)\left(\frac{1}{\hat{\mathfrak{D}}(\vec{l})} - 1 - \frac{\hat{l}_x\hat{l}_y}{l^2}\right) - \frac{2\hat{F}(\vec{l})\hat{l}_x\hat{l}_y}{\hat{\mathfrak{D}}(\vec{l})} + \frac{\hat{l}_x\hat{l}_y}{l^2} + \frac{2\hat{F}_x(\vec{l})}{\hat{\mathfrak{D}}(\vec{l})}\hat{l}_y^2\right]. \tag{B42}$$

Again we observe that $\Delta^\alpha(D)$ cannot be expanded in power series of D^2 for the same reason as we discussed in the case of $\eta(D)$.

The evaluation of the nonlinear shear viscosity effects thus involves the calculation of several integrals which when expressed in terms of spherical polar coordinates are of the form

$$\delta_j = \int_0^\infty\int_0^\pi\int_0^{2\pi} r^2\sin\theta f_j(\vec{r})\,dr\,d\theta\,d\phi, \tag{B43}$$

where $j = 0, x, y, z$, with

$$\Delta^j = \frac{k_B T}{(2\pi)^3}\left(\frac{\rho D}{\eta}\right)^{3/2}\delta_j, \quad j = x, y, z \tag{B44}$$

and

$$\Delta\eta = \frac{k_B T}{(2\pi)^3} D^{1/2}\left(\frac{\rho}{\eta}\right)^{3/2}\delta_0 \tag{B45}$$

as given in Eqs. (B38), (B40), (B41), and (B42).

Each of the integrands involve the functions $\hat{F}_x$ and $\hat{F}$ defined through the integrals in Eqs. (B36) and (B37). As one is unable to evaluate $\hat{F}_x$ and $\hat{F}$ analytically [except for certain special angles ($\phi = 0$, π or 2π)] one must evaluate both of these functions and the δ's numerically, which was done with the use of the CDC 6400. The procedure used is the following: The integral corresponding to a given δ_j was divided into several parts:

$$\delta_j = \delta_{j,0} + \delta_{j,1} + \delta_{j,2}, \tag{B46}$$

where

$$\delta_{j,0} = \int_0^{R_0}\int_0^\pi\int_0^{2\pi} r^2\sin\theta f_j(\vec{r})\,dr\,d\theta\,d\phi, \tag{B47}$$

$$\delta_{j,1} = \int_{R_0}^{R_1}\int_0^\pi\int_0^{2\pi} r^2\sin\theta f_j(\vec{r})\,dr\,d\theta\,d\phi, \tag{B48}$$

$$\delta_{j,2} = \int_{R_1}^\infty\int_0^\pi\int_0^{2\pi} r^2\sin\theta f_j(\vec{r})\,dr\,d\theta\,d\phi, \tag{B49}$$

where R_0 and R_1 were chosen to be 10^{-4} and 2, respectively.

The functions $\hat{F}_x$ and $\hat{F}$ were evaluated numerically using Simpson's method. It was possible by a combination of numerical and analytical methods to then show that $\delta_{j,0}=O(R_0)$ and hence is negligibly small. The integrals $\delta_{j,1}$ were evaluated on the computer using Simpson's method, with the results

$$\delta_{0,1}=-1.26,\quad \delta_{x,1}=-0.24,\qquad \delta_{y,1}=+1.23,\quad \delta_{z,1}=-1.07. \tag{B50}$$

The evaluation of the remaining contribution $\delta_{j,2}$ was done analytically, making use of a Taylor-series expansion for $\hat{F}_x$ and $\hat{F}$ (and hence f_j) in powers of r^{-2}, about the point $r^{-1}=0$. Although one could not prove that these Taylor-series expansions converged, comparison with the numerical calculation of $\hat{F}_x$ and $\hat{F}$ indicated that the first term in such an expansion is a very good approximation to these functions. Hence $\delta_{j,2}$ was evaluated analytically, keeping the first two leading terms in the expansion for $\hat{F}$ and $\hat{F}_x$, with the results

$$\delta_{0,2}=-1.2\times10^{-4},\quad \delta_{x,2}=-1.8\times10^{-1},\qquad \delta_{y,2}=+1.5\times10^{-2},\quad \delta_{z,2}=-6.0\times10^{-2}. \tag{B51}$$

The resultant values for Δ_j are thus found to be

$$\begin{aligned}\Delta\eta &\cong -5.1\times10^{-3}k_BT(\rho/\eta)^{3/2}D^{1/2},\\ \Delta^x &\cong -1.7\times10^{-3}k_BT(\rho D/\eta)^{3/2},\\ \Delta^y &\cong +5.0\times10^{-3}k_BT(\rho D/\eta)^{3/2},\\ \Delta^z &\cong -4.6\times10^{-3}k_BT(\rho D/\eta)^{3/2},\end{aligned} \tag{B52}$$

with an estimated error of about 10% for the above numerical coefficients.

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[12]A similar transformation has been also introduced by Robertson (Ref. 3) for his projection operator.

[13]This type of projection operator, however, does not remove those long-time memory effects which arise from nonlinear coupling among hydrodynamic modes, as has been discussed recently (Refs. 6 and 8). In this section dealing with a general formalism, we disregard this aspect of the problem. In actual applications, of course, these long-time contributions must be considered explicitly. See Sec. III.

[14]For normal stress effects, see, e.g., the article by H. Markovitz, in *Rheology, Theory and Applications*, edited by F. R. Eirich, (Academic, New York, 1967) Vol. 4.

[15]In the expression next to (B.1), in Ref. 6, $(\partial/\partial\vec{r}_i)\vec{v}(\vec{r}_i)$ in fact is meant to be $[\partial\vec{v}(\vec{r}_i)/\partial\vec{r}_i]$.

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