Mode-coupling contributions to the nonlinear shear viscosity

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(Received 22 March 1985)

Three different results for the nonanalytic dependence of the shear viscosity on shear rate have been given. This discrepancy is resolved by an independent calculation based on the nonlinear Navier-Stokes-Langevin equations. The relationship to previous calculations and reasons for the differences are described.

More than a decade ago Kawasaki and Gunton[1] showed that the nonlinear response of a fluid under shear is nonanalytic in the shear rate. Their calculation involved two components. First, the nonequilibrium statistical mechanics for linear response was extended to states far from equilibrium to identify time correlation function expressions for transport coefficients. Next, these expressions were evaluated for shear flow using a nonequilibrium version of Kadanoff-Swift mode-coupling theory.[2] They found that for an asymptotically small shear rate, the nonlinear shear viscosity has the form

$$\eta(a) \rightarrow \eta_0 + \eta_1 a^{1/2} , \tag{1}$$

where a is the magnitude of the shear rate and η_0 is the shear viscosity for a Newtonian fluid. The coefficient η_1 was expressed in terms of several mode-coupling integrals that were evaluated numerically. Subsequently, Yamada and Kawasaki[3] formulated the problem in terms of a Fokker-Planck equation associated with the nonlinear Navier-Stokes-Langevin equations. Approximations were introduced that amount to replacing the nonlinear Fokker-Planck equation by the corresponding equation linearized around the local equilibrium state. A time correlation function expression for the shear viscosity was obtained, similar to that of Kawasaki and Gunton. The numerical value for η_1 determined by Yamada and Kawasaki differs from that of Kawasaki and Gunton. Both calculations were limited to mode-coupling effects in an incompressible fluid. Several years later, Ernst *et al.*[4] used a kinetic theory in the "ring approximation" to discuss nonlinear transport in a compressible fluid. Although their derivation is limited to a low density gas, the final results are expressed in terms of the thermodynamic parameters for a general fluid. They obtained additional contributions to η_1 due to compressibility effects, but even in the incompressible limit their value differs from both previous results.

These discrepancies might not be noteworthy except for recent nonequilibrium computer-simulation results[5] that suggest still different values for η_1, several orders of magnitude larger. Although some related mechanism may be responsible for this striking difference between theory and computer simulation,[6] it remains to remove the possibility of a serious error in the existing theoretical mode-coupling calculations. In addition, there is interest in a possible relationship of the value of η_1 to the mode-coupling amplitude of long-time tails for equilibrium fluctuations.[7] Reliable theoretical values of η_1 are required to test the proposed relationship. The objective here is to report the results of a direct calculation of η_1 from the nonlinear Navier-Stokes-Langevin equations for a compressible fluid in the two-mode-coupling approximation. Complete numerical agreement with Ernst *et al.* is obtained. Furthermore, in the incompressible-fluid limit the analytic expressions of Ernst *et al.*, Yamada and Kawasaki, and those reported here, are all found to be equivalent. It is concluded that the difference of Yamada and Kawasaki's value is due to an inaccuracy of the numerical integration. It is also suggested that higher-order mode-coupling effects retained by Kawasaki and Gunton should be neglected for self-consistency. In this case their results reduce to those of Yamada and Kawasaki.

The nonlinear Navier-Stokes-Langevin equations[8] are a model of the microscopic local conservation equations for the mass, energy, and momentum densities. The fluxes in these equations can be separated into Euler and dissipative parts in analogy to the macroscopic hydrodynamic fluxes. The remaining "fast" degrees of freedom at the microscopic level are represented by an additional random component with Gaussian-Markovian statistics. For example, the Euler contributions to the microscopic stress tensor are identified by a Gallilean transformation to the local rest frame,

$$\hat{t}_{ij}(\mathbf{r}) = \hat{\rho}(\mathbf{r})\hat{v}_i(\mathbf{r})\hat{v}_j(\mathbf{r}) + \hat{t}'_{ij}(\mathbf{r}) , \tag{2}$$

where $\hat{t}_{ij}$ is the stress tensor and $\hat{t}'_{ij}$ is the same quantity referred to the local rest frame. The latter is defined by the velocity field $\hat{v}_i = \hat{\rho}^{-1}\hat{p}_i$, where $\hat{\mathbf{p}}$ is the momentum density and $\hat{\rho}$ is the mass density. The caret over these quantities is used to differentiate these phase functions from their corresponding hydrodynamic (macroscopic) variables. Next, the stress tensor in the local rest frame is further separated into a local pressure $\hat{p}$, dissipative part $\hat{t}^*_{ij}$, and a random component $\hat{t}^R_{ij}$. Then Eq. (2) becomes

$$\hat{t}_{ij}(\mathbf{r}) = \hat{\rho}(\mathbf{r})\hat{v}_i(\mathbf{r})\hat{v}_j(\mathbf{r}) + \hat{p}(\mathbf{r})\delta_{ij} + \hat{t}^*_{ij}(\mathbf{r}) + \hat{t}^R_{ij}(\mathbf{r}) .$$

The macroscopic stress tensor has a form similar to (3) in terms of the hydrodynamic variables,

$$t_{ij}(\mathbf{r}) \equiv \langle \hat{t}_{ij}(\mathbf{r}) \rangle \tag{3}$$

$$= \rho(\mathbf{r}) U_i(\mathbf{r}) U_j(\mathbf{r}) + p(\mathbf{r})\delta_{ij} + t^*_{ij}(\mathbf{r}) , \tag{4}$$

where p and ρ are the macroscopic pressure and density, and $\mathbf{U}(\mathbf{r})$ is the macroscopic flow velocity. The brackets denote an average over the appropriate nonequilibrium state. Finally, t^*_{ij} is the irreversible part of the macroscopic stress tensor.

For uniform shear flow, the macroscopic velocity is taken along the x direction with constant gradient along the y axis,

$$U_x = ay, \quad U_y = U_z = 0 . \tag{5}$$

The nonlinear shear viscosity $\eta(a)$ is defined by

$$t_{xy}^* = -\eta(a)a \ . \tag{6}$$

An expression for $\eta(a)$ now follows from Eqs. (3) and (4),

$$\eta(a)a = -\langle \hat{t}_{xy}^* \rangle + \rho U_x U_y - \langle \hat{\rho}\hat{v}_x\hat{v}_y \rangle \ , \tag{7}$$

where the average of the random component has been defined as zero. The expression (7) is quite general. To calculate the right side of this equation, $\hat{t}_{xy}^*$ is defined in the Navier-Stokes limit (small spatial variations), by analogy with Newton's viscosity law,

$$\hat{t}_{xy}^* = -\eta_B \left[\frac{\partial v_x}{\partial y} + \frac{\partial v_y}{\partial x} \right] , \tag{8}$$

where the bare viscosity η_B has been distinguished from the true viscosity η. Equations (8) and (3), together with a similar decomposition of the energy flux, define the Navier-Stokes-Langevin equations (further details are given in Ref. 8). These are a closed set of equations from which all quantities in (7) can be calculated. The deviations of the microscopic variables from their macroscopic values are due to thermal fluctuations. To the extent that these are small, it is reasonable to simplify (7) further by retaining only such terms to quadratic order. The result is

$$\eta(a) = \eta_B - \rho \langle \delta v_x(\mathbf{r}) \delta v_y(\mathbf{r}) \rangle \ . \tag{9}$$

For self-consistency, the correlation function on the right side of (9) should be calculated from the Navier-Stokes-Langevin equations to linear order in these deviations. Equation (9) will be referred to as the two-mode-coupling approximation.

To evaluate the correlation function in (9) it is convenient to use the equivalent Fourier representation

$$\eta(a) - \eta_B = \rho V^{-1} \sum_{\mathbf{k}} \langle v_x(\mathbf{k}) v_y(-\mathbf{k}) \rangle \ , \tag{10}$$

where $v_i(\mathbf{k})$ is the Fourier transform of $\delta v_i(\mathbf{r})$, and V is the volume. Correlation functions of the type in (10) have been evaluated elsewhere[9] for the shear-flow problem in terms of the nonequilibrium hydrodynamic modes. At small shear rates there are dominant contributions from coupling of shear modes and coupling of sound modes. In the notation of Ref. 4, Eq. (1) is obtained with

$$\eta_1 = k_B T \left[\frac{M^{+-}}{(2D_s)^{3/2}} + \frac{M^{\eta\eta}}{(2D_\eta)^{3/2}} \right] \ . \tag{11}$$

Here T is the temperature, k_B is Boltzmann's constant, D_η is the thermal diffusivity, D_s is the sound-damping constant, and M^{+-} and $M^{\eta\eta}$ are dimensionless constants associated with mode-coupling integrals for sound and shear modes, respectively. A comparison of the values obtained for these constants is given in Table I. (M^{+-} applies only for a compressible fluid and is not obtained in Refs. 1 and 3; also included in Table I are some other mode-coupling constants of Ref. 4). The agreement of the present results with those of Ernst *et al.* are within the bounds of the numerical accuracy.

To explain the discrepancy with Yamada and Kawasaki, it is first noted that their definition of the shear viscosity is the same as Eq. (10). Their procedure is then to relate the nonequilibrium average to a local equilibrium average, neglecting nonlinear terms in their Fokker-Planck equation, to obtain the form

$$\eta(a) = \rho^2 (k_B T V)^{-1} \sum_{\mathbf{k}} \sum_{\mathbf{k}'} \int_0^\infty dt \langle v_x(\mathbf{k},t) v_y(-\mathbf{k},t) v_x(\mathbf{k}') v_y(-\mathbf{k}') \rangle_{l,c} \tag{12}$$

where $v_\alpha(\mathbf{k},t)$ are solutions to the (incompressible) Navier-Stokes equations lineraized around shear flow. The brackets $\langle \cdots \rangle_{l,c}$ denote a Gaussian local equilibrium distribution and the subscript c means a cumulant with $v_x(\mathbf{k})v_y(-\mathbf{k})$ regarded as a single unit. Although the form of (12) is quite different from (10), the approximations involved are the same as those of the Langevin calculation given here. In fact, it has been verified in detail that the integrals required to evaluate η_1 in Yamada and Kawasaki's expression [Eq. (B.8) of Ref. 3] is equivalent to that of the present work. Furthermore, the integral expressions of Ernst *et al.* for the mode-coupling coefficients in Table I (Appendix B of Ref. 4) have also been shown to be equivalent to those reported here. Consequently, the Fokker-Planck, kinetic theory, and Langevin methods all lead to the same result, and differences are attributed to inaccuracies in numerical integration.

The expression for the shear viscosity given by Kawasaki and Gunton also starts with the two-mode-coupling approximation (10) but their transformation to a local equilibrium

TABLE I. Comparison of mode-coupling coefficients $\times 10^2$.

	$M^{\eta\eta}$	M^{+-}	$M^{(0)}$	$M^{(1)}$	$M^{(2)}$
Kawasaki and Gunton (Ref. 1)	−1.4	...	...	...	...
Yamada and Kawasaki (Ref. 3)	+0.86	...	...	...	...
Ernst *et al.* (Ref. 4)	−0.26	−0.406	0	+0.507	−0.766
Present work	−0.256	−0.417	0	+0.510	−0.766

average is somewhat more complex than (12),

$$\eta(a)=a^{-1}\rho V^{-1}\sum_{\mathbf{k}}\left\langle v_x(\mathbf{k})v_y(-\mathbf{k})\left[1-\exp\left(-a\rho(k_B T)^{-1}\sum_{\mathbf{k}'}\int_0^\infty dt\, v_x(\mathbf{k},-t)v_y(-\mathbf{k},-t)\right)\right]\right\rangle_{l,c} . \tag{13}$$

The time-reversed equations for $v_i(\mathbf{k},-t)$ have the same form as those for $v_i(\mathbf{k},t)$ in Yamada and Kawaski, except with the sign of the shear rate reversed. If the exponential in (13) is expanded to linear order, the result is the same as (12) [since $\eta(a)=\eta(|a|)$]. Consequently, differences in the predicted values of η_1 from (13) can be attributed to higher-order terms in the expansion of the exponential. Since the initial expression (10) is already limited to a two-mode approximation, it does not appear self-consistent to retain such higher-order mode-coupling terms in (13).

A unique feature of the Langevin method applied here is its simplicity. Although the results are equivalent to those of Refs. 1 and 3, the latter ultimately have to evaluate a local equilibrium time correlation function of four velocity fields. Here the nonequilibrium time-independent correlation function of two velocity fields [Eq. (10)], is evaluated directly. Such static correlation functions obey simple linear equations that follow directly from the Langevin equations. Beyond the two-mode approximation this relative simplicity is less apparent.

The authors are indebted to Bob Coldwell for advice and assistance with the numerical integration. This research was supported by National Science Foundation Grant No. CHE-84-11932.

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