Nonequilibrium Molecular-Dynamics Simulation of Couette Flow in Two-Dimensional Fluids

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Theory predicts that in two dimensions, the Navier-Stokes transport coefficients diverge. The results of molecular-dynamics simulations of viscous flow for a two-dimensional fluid are reported. The results suggest that, at *low* shear rate, instabilities in the flow screen the predicted logarithmic divergence of viscosity with respect to strain rate. When a Maxwell demon is used to suppress these instabilities, the computed viscosity is consistent with the divergent behavior predicted theoretically.

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Ever since the discovery of "long-time tails" by Alder and Wainwright," there has been considerable interest in the nature of hydrodynamics in two dimensions. Alder and Wainwright[1] used equilibrium molecular dynamics to show that the Green-Kubo time correlation function for self-diffusion $Z(t)$, the velocity autocorrelation function

$$Z(t)=\tfrac{1}{3}\langle\vec{V}_i(t)\cdot\vec{V}_i(0)\rangle, \quad (1)$$

decays very slowly at long time. A combination of theory and simulation[1,2] lead to the prediction that

$$\lim_{t\to\infty} Z(t)\sim t^{-d/2}, \quad (2)$$

where d is the dimensionality of the system ($d>1$). Since the limiting zero-frequency self-diffusion coefficient is the integral of $Z(t)$, the self-diffusion coefficient must diverge in two-dimensional (2D) systems !

As has been pointed out many times, however,[2] there is an inconsistency in this argument. In order to derive the Green-Kubo relation

$$D=\int_0^\infty dt\, Z(t), \quad (3)$$

one must assume that the relevant diffusive processes can be described by the linearized Navier-Stokes equation. Starting with this assumption one then derives the Green-Kubo formula (3). Subsequently kinetic theory[2] predicts the existence of long-time tails which ultimately lead to the nonexistence of linearized hydrodynamics ! The way out of this dilemma is to realize that the existence of t^{-1} long-time tails shows that in 2D the derivations of Green-Kubo relations are invalid. One cannot therefore trust the Green-Kubo formulas to calculate Navier-Stokes transport coefficients in two dimensions. At the present time there is only one other technique available. It is nonequilibrium molecular dynamics.

In this paper we use nonequilibrium molecular dynamics to calculate the shear viscosity of a two-dimensional fluid at zero frequency and zero wave vector. This technique has been discussed many times.[3] As the name suggests, the shear viscosity η is calculated by taking the ratio of stress $-P_{xy}$ to strain rate $\gamma=\partial u_x/\partial y$. The computed viscosity is observed to be a function of shear rate. Kawasaki and Gunton (KG)[4] were the first to show that "long-time-tail" processes are expected to lead to nonanalytic, constitutive relations for the nonlinear shear viscosity $\eta(\gamma)$. In two dimensions they predicted that[4]

$$\lim_{\gamma\to 0}\eta(\gamma)\sim\log(\gamma) \quad (4)$$

and

$$\lim_{\gamma\to 0} p(\gamma)\sim\gamma\log\gamma. \quad (5)$$

In 1980 one of us[5] reported computer simulations for small ($N=32$, 50, 98) two-dimensional systems showing that (4) and (5) were indeed consistent with the numerical results. As pointed out in that paper,[5] there was a large dependence of the results upon the size of the system studied. Recently there have been improvements in simulation techniques[6,7] which allow us to simulate much larger systems, $N=896$, 3584. As we shall see the $N=896$ and 3584 results are essentially identical.

The system studied was N soft disks with

$$\varphi(r)=\epsilon(\sigma/r)^{12} \quad (6)$$

at a temperature $kT/\epsilon=1$ and a series of densities $\rho\sigma^2=0.96$, 0.9238, 0.6928. The truncation distance is 1.5σ. The highest density is very close to the freezing transition[8] ($\rho_f\sigma^2=0.986\pm0.001$, $\rho_m\sigma^2=1.007\pm0.001$). Figure 1 shows the reduced viscosity η^* as a function of the common logarithm of the reduced strain rate γ^* (= $\partial u_x*/$

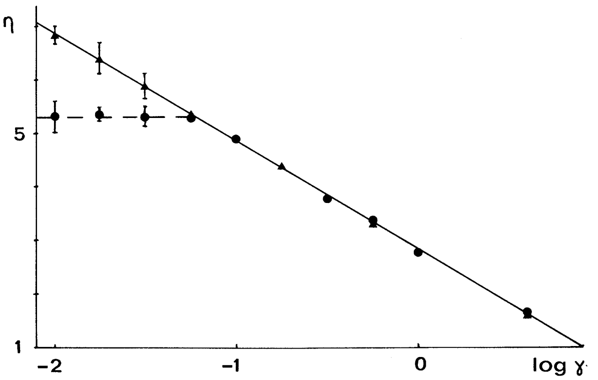

FIG. 1. The effective viscosity as a function of $\log_{10}\gamma$ ($N = 896$, $\rho\sigma^2 = 0.96$). Dots depict the results of Doll's-tensor simulations. Note the turnover due to kink instabilities for $\gamma \lesssim 0.1$. Triangles depict the results of constrained Doll's-tensor simulations. These two sets of results show that the KG logarithmic divergence is only obtained for steady, planar Couette flow. All quantities are expressed in units of m, σ, ϵ.

∂y^*) for the high-density state with $N = 896$. At low γ^* we can see two sets of results. One is essentially flat—the "turnover"—and the other follows a logarithmic variation with γ. The turnover is the naturally occurring result. We will explain later how the extended logarithmic result was obtained. At lower densities (Figs. 2 and 3) we see the same qualitative features. At high γ the viscosity is consistent with the KG functional form but then as we lower γ we enter a turnover regime.

The critical γ at which the transition occurs increases as we decrease the density until at our lowest density the turnover regime extends over much of the accessible range of strain rates. Also in Fig. 3 we can see that within the statistical uncertainties, the viscosity is independent of γ in the turnover regime.

In Fig. 2 we see a comparison of Doll's-tensor simulations[7] with the homogeneous shear technique where the flow is driven solely by the periodic image convention. We see that the two sets of calculations yield the same results in both the logarithmic and the turnover regimes. In Fig. 2 we can see that in both regimes the results are independent of the system size. The results for $N = 896$ and 3584 are statistically indistinguishable.

What are we to make of this extraordinarily sharp transition from the logarithmic (KG) regime to this apparently flat turnover regime?

One of the extraordinary features of the KG prediction is that at sufficiently low strain rates two-dimensional fluids become negatively dilatant ($\gamma \log\gamma$ is nonmonotonic). Thus the KG theory predicts that in a *finite* range of shear rates about the equilibrium state (= 0), shearing two-dimensional fluids should peak more efficiently than they do at equilibrium!

A thermodynamic argument can be produced showing that such a situation is impossible. For a nonequilibrium steady state[9] we have

$$dE = T\,ds - p\,dV + \zeta\,d\gamma, \tag{7}$$

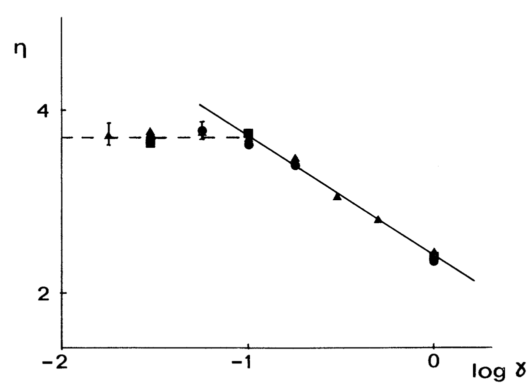

FIG. 2. $\eta(\gamma)$ as a function of $\log_{10}\gamma$ ($\rho\sigma^2 = 0.9238$). Triangles depict homogeneous shear, $N = 896$; dots depict homogeneous shear, $N = 3584$; squares depict Doll's-tensor shear, $N = 896$. All calculations agree with each other within statistical uncertainties. There is no N dependence or method dependence. The unstable turnover occurs at slightly larger γ than for $\rho\sigma^2 = 0.96$ (Fig. 1).

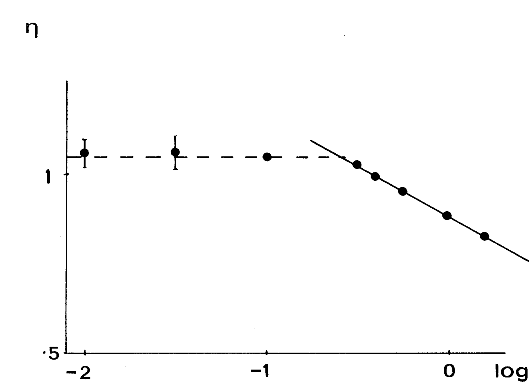

FIG. 3. $\eta(\gamma)$ as a function of $\log_{10}\gamma$ for Doll's-tensor simulations of 896 soft disks at $\rho\sigma^2 = 0.6928$. The unstable turnover region now extends to large γ (= 0.25), making it difficult to observe the logarithmic regime. Note the virtual independence of viscosity upon γ in the turnover regime.

where $\zeta = \zeta(T, V, \gamma)$ measures the dependence of internal energy E upon the magnitude of the strain rate, γ. It is easily shown that

$$\partial\zeta/\partial\gamma \geq 0. \tag{8}$$

If this thermodynamics is to reduce to equilibrium thermodynamics then $\zeta(T, V, \gamma=0)=0$. Combining this with (6) we see that for all state points,

$$\zeta(T, V, \gamma) \geq 0. \tag{9}$$

It is easy to see that the KG shear dilatancy (5) violates this thermodynamic stability requirement (9). This follows since ζ can be evaluated from the equation

$$\zeta(T, V, \gamma) = \int_v^\infty \frac{\partial p}{\partial \gamma}(V', T, \gamma)\,dV' + (\text{ideal gas term}). \tag{10}$$

If the KG constitutive reaction (5) is substituted into (10) we find that KG predicts $\zeta(\gamma) \sim \log\gamma$. For sufficiently small strain rates ζ can become negative. We believe that this thermodynamic instability is the explanation for the turnover from the logarithmic (KG) regime.

In Fig. 4 we show shear dilatancy as a function of γ for the intermediate density. We see that when the system appears to be entering the negative-dilatancy region, the turnover occurs. The pressure suddenly becomes essentially independent of shear rate, taking essentially the same

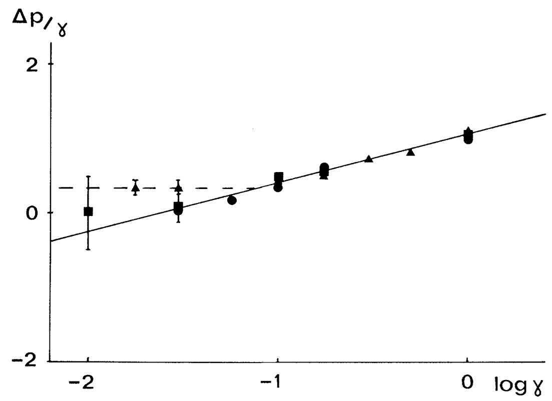

FIG. 4. Shear dilatancy $\Delta p(\gamma)/\gamma$ as a function of $\log_{10}\gamma$ for $\rho\sigma^2 = 0.9238$. The symbols have the same meaning as in Fig. 2. If the data followed KG to equilibrium then negative dilatancy would be observed for $\gamma \lesssim 0.03$. This is not seen. At an apparently higher strain rate $\gamma \sim 0.1$, kink instabilities screen the KG result and all properties become essentially independent of γ.

value as the equilibrium pressure.

Thermodynamics does not of course provide us with a microscopic understanding of exactly what processes are responsible for the turnover. The turnover region is not easy to simulate. Fluctuations are large but not simply because the strain rate is small.

For a variety of reasons we believe that the turnover was caused by transverse momentum currents. To test this hypothesis we constructed a Maxwell demon to straighten out these suspected momentum kinks. This demon was constructed in the following manner.

If

$$\dot{\vec{q}}_i = \vec{p}_i/m + \vec{q}_i \cdot \nabla\vec{u}, \tag{11}$$

then the momentum $\{\vec{p}_i\}$ defines a periodic peculiar momentum field which can by synthesized in a Fourier series:

$$\vec{p}(\vec{k}_{\vec{n}}) = \sum \vec{p}_i \exp(i\vec{k}_{\vec{n}} \cdot \vec{q}_i), \tag{12}$$

where

$$\vec{k}_{\vec{n}} = 2\pi/L(n_x, n_y); \quad n_x, n_y = 0, \pm 1, \pm 2, \ldots . \tag{13}$$

Our Maxwell demon ensured that the first three transverse modes[10] of the momentum field were all zero. That is for $\vec{n}=(0,1)$, $(0,2)$, $(0,3)$ and $(1,0)$, $(2,0)$, $(3,0)$, it forced $\vec{p}(\vec{k}_n)=0$. This is accomplished by adding the appropriate canceling Fourier harmonics to the particle momenta. Because of computational expense this was done once every ten time steps during the course of the simulation.

As can be seen in Fig. 1 the effect of this straightening is dramatic. It totally removes the turnover regime, with the result that over $2\frac{1}{2}$ decades of strain rate the viscosity accurately tracks the KG functional form. The Maxwell demon has no effect in the preexisting KG regime. The velocity profile was already linear so that the straightening had no effect.

In the preexisting turnover regime the thermodynamic instability manifests itself in convective cells destroying the linearity of the velocity profile. Evidently the number and amplitude of these cells vary with γ in such a way as to prevent the thermodynamic strain rate potential, ζ, from ever becoming negative.

We have presented results which show clearly that steady planar Couette flow in two dimensions becomes unstable at sufficiently *small* strain rates. This instability screens the predicted logarithmic divergence of the two-dimensional viscosity with respect to strain rate. We believe that these effects invalidate existing theories of Navier-Stokes transport in two dimensions. It has long been known that close to equilibrium two-dimensional systems are highly nonlinear. Evidently this nonlinearity is so strong that not only do the usual mode couplings occur between linear, bilinear, ... variables of the same wave vector, but couplings also occur between phase variables of different wave vectors. The latter processes are responsible for the excitation of convective cells in the flow.

Note added.—A partial account of this work will be reported in Physics Today.[11]

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