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Shear viscosity via global control of spatiotemporal chaos in two-dimensional isoenergetic dense fluids

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We control the chaotic large-scale spatiotemporal fluctuations inherent in large-scale two-dimensional nonequilibrium flows by using global ergostats. We use isoenergetic nonequilibrium molecular dynamics to characterize the size dependence of both the shear viscosity and the largest Lyapunov exponent for two-dimensional globally controlled dense periodic fluids. Though uncontrolled fluctuations in such flows have often been thought to lead to divergent transport coefficients in two dimensions, our numerical evidence shows instead that the large-system "thermodynamic limit" can be extended to a convergent homogeneous "hydrodynamic limit" away from equilibrium, with finite transport coefficients, even in two dimensions.

I. INTRODUCTION

It has been well known for over 20 years [1,2] that (the Green-Kubo expressions for the) transport coefficients diverge in two space dimensions. It has, similarly, but much less convincingly, been suggested [3], based on the logarithmic divergence of the mean-squared displacement [4], that two-dimensional *solids* do not exist. Both these "divergences" are special to two-dimensional systems. They reflect the relative importance of fluctuations, relative to surface effects, in that case [4]. In two dimensions both these effects contribute terms of order $N^{-1/2}$ to N-body intensive properties.

Despite the theoretically based divergences, many [5] varied two-dimensional fluid- and solid-phase many-body simulations have been carried out, with no apparent difficulties. We recently suggested [6] that homogeneous global constraint forces can be used to eliminate any such divergent fluctuations. Our investigations show that constraint forces can be used to construct a "hydrodynamic limit" for large-size nonequilibrium systems of any dimensionality provided that the corresponding nonequilibrium state is either stationary or periodic in time. The existence of these nonequilibrium limiting states is analogous to the extensivity of the free energies in the large-system equilibrium "thermodynamic limit." The limiting nonequilibrium states are characterized by finite shear stresses and heat fluxes in addition to the usual equilibrium state variables.

The many-body systems treated by molecular dynamics are typically chaotic, and characterized by a spectrum of Lyapunov exponents [6,7]. In the nonequilibrium case it is essential to apply "feedback" or "control theory" to constrain these large chaotic systems to steady nonequilibrium states. Thus the molecular chaos inherent in the many-body problem of molecular dynamics has long been controlled, with thermostats, barostats, ergostats, etc. More recently, control of spatiotemporal chaos has caught the attention of specialists in nonlinear dynamics. It is possible that these two fields will become more closely coordinated in the future. Two-dimensional viscous flows, like those we consider here, have been simulated for more than 20 years [4,8]. However, no clear consensus has emerged regarding the size dependence of the results [8]. In part, this is because previous investigators have examined only a few similar system sizes. Thus the range of $\ln N$ may not have been sufficiently large to confirm or deny a logarithmic dependence; in part, the lack of consensus reflects the relatively large fluctuations found in two-dimensional flows. With the gradual incremental increases in computer speed and capacity, we are now able to carry out relatively long and accurate simulations with as many as a quarter million particles. In two dimensions, accuracy requires relatively long runs. Thus we have reinvestigated the two-dimensional shear problem. We describe the simulations and the results here, after first describing our own understanding of the fluctuation-based difficulties associated with two-dimensional systems.

We set out to generate convincing numerical evidence for the presence or absence of the number dependence of two-dimensional viscosities in the low-strain-rate regime where the distraction of "string" phases is not present. Accordingly, here we study the dependence of two-dimensional dense-fluid shear viscosity on system size alone, using a short-ranged specially smooth repulsive potential, and considering systems ranging from 64 up to 264 196 particles at a strain rate small enough to avoid string-phase anomalies but still large enough to minimize statistical fluctuations.

The method used, constant-energy nonequilibrium molecular dynamics with shearing periodic boundary

conditions, is not new [9]. It is a reliable tool for generating estimates of the shear and bulk [10] viscosities as well as the thermal conductivity [11]. We integrate the nonequilibrium equations of motion using the classic fourth-order Runge-Kutta method, for a special short-ranged potential [7] with a very smooth cutoff, in order to eliminate numerical errors. We discuss the "divergence" problem further in Sec. II. We give the details of our model dense-fluid system, together with Enskog's predictions of its shear behavior, in Secs. III and IV. Our numerical shear-viscosity results are then presented and discussed in Secs. V and VI.

II. DIVERGENCE OF TRANSPORT COEFFICIENTS IN TWO DIMENSIONS

Green and Kubo's linear response theory, applied to classical fluids, suggests that the linear transport coefficients can be written as time integrals of equilibrium two-time correlation functions. For the case of interest to us here, the shear viscosity η, that coefficient is given by the theory in terms of a shear-stress autocorrelation integral

$$\eta(t \to \infty) = (V/kT)$$
$$\times \int_0^{t \to \infty} \langle P_{xy}(0)P_{xy}(t') \rangle_{\text{equilibrium}} dt' \, ,$$

where the angular brackets indicate a canonical-ensemble average. Intuition, hydrodynamic mode-coupling theory, as well as few-body low-density kinetic theory, all lead to the same plausible conclusion: fluctuation amplitudes decay as $1/t$ in two dimensions, leading to divergent autocorrelation integrals $\eta(t \to \infty) \approx \ln t$. This result suggests a much less plausible conclusion: transport coefficients diverge in two dimensions. The results we present here, together with recent independent calculations carried out by Gravina, Ciccotti, and Holian [12], indicate that this divergence argument has some exceptions.

What *has* been shown theoretically is that the Green-Kubo expressions probably do diverge for infinitely large two-dimensional systems so long as boundaries are *not* taken into account. There is considerable computer-based support for this view in diffusive systems [2]. What might the demonstration of divergence for infinitely large equilibrium systems mean for finite fluids and solids, away from equilibrium and *with* boundaries? For shear viscosity, such a divergence might imply the lack of a limiting linear stress versus strain rate relation for a two-dimensional system of any size. Alternatively, it might indicate the presence of a linear relation linking stress and strain rate, but with a number-dependent divergence (probably logarithmic) of the measured transport coefficient $\eta(N)$ as the system size is increased.

In either case it is evident that the "divergence" somehow reflects the enhanced relative importance of fluctuations in two-dimensional systems. The fluctuations are apparent in the dynamics of crystal lattices. There the rms spatial fluctuation of a solid-phase atom can be evaluated quantitatively, in terms of the crystal's frequency spectrum. If this fluctuation is studied as a function of system size, an interesting qualitative dependence on the dimensionality of the system is observed [4]. For two-dimensional periodic crystals a plot of $\langle \delta r^2 \rangle$ versus $\ln N$ is a straight line with a well-understood slope [4]. In three dimensions $\langle \delta r^2 \rangle$ varies instead linearly with $N^{-1/3}$, approaching a finite limit as N increases to infinity. Why is there a qualitative dependence on dimensionality [divergence in two dimensions and convergence in three] rather than just a quantitative one?

Thermal mass, momentum, and energy fluctuations in portions of a large two-dimensional system can confidently be expected to obey the central limit theorem. That is, in a small region containing $N \gg 1$ atoms the fluctuation amplitudes for extensive quantities, proportional to $N^{+1/2}$, are of the same order as are the surface contributions of the region. The local fluctuations in two-dimensional systems have to be maintained by corresponding surface fluxes of mass, momentum, and energy. In three dimensions mass, momentum, and energy still fluctuate locally, but these thermal fluctuations, of order $N^{+1/2}$, are negligible, for sufficiently large N, relative to the surface area of a three-dimensional region, which grows as $N^{+2/3}$ rather than as $N^{+1/2}$.

The large-system Reynolds number for dense-fluid flows provides a dramatic example of the relative importance of fluctuations in two dimensions. For a system with a characteristic velocity v, size L, and kinematic viscosity (η/ρ), the Reynolds number is $\text{Re} \equiv vL/(\eta/\rho)$. For thermal fluctuations with N microscopic particles at a temperature T, the uncertainty in the center-of-mass velocity Δv is of order $(kT/mN)^{1/2}$. With an atomistic particle size σ, the two-dimensional system size is of order $L \approx N^{1/2} \sigma$. Using an order-of-magnitude estimate for the dense-fluid kinematic viscosity $(\eta/\rho) \approx (kT/m)^{1/2}\sigma$, establishes that the uncertainty in the Reynolds number remains of order unity, even in the large-system limit:

$$\Delta \text{Re}_{2D} = \Delta v L/(\eta/\rho)$$
$$\approx (kT/mN)^{1/2}N^{1/2}\sigma/[(kT/m)^{1/2}\sigma] \equiv 1 \, .$$

Thus there is no possibility that the flow of two-dimensional fluids at very small Reynolds numbers can be treated accurately by ordinary hydrodynamics, because hydrodynamics ignores fluctuations. In three dimensions fluctuations are less important, and these difficulties are not so serious. There the center-of-mass contribution to the Reynolds number decreases $(\Delta \text{Re}_{3D} \propto N^{-1/6})$ as the system size increases.

Even in three dimensions, where the large-system Reynolds number *is* well defined, sufficiently large regions do become turbulent and in large volumes fluctuations certainly decay slowly. Consider, for example, a 1-m cube of water. The cube exhibits turbulence at boundary velocities of order one mm/s. The time required for fluctuations to dissipate, according to conventional hydrodynamics, is on the order of a year for the cube. Brush [13] has chronicled Clausius's description of the small scale velocity fluctuations ("the kind of motion we call heat"). In what sense can this perpetual thermal motion of a three-dimensional cube of water be said to obey conventional hydrodynamics? At least a spatial average, and

possibly also a time, or ensemble, average, would be required to make a correspondence.

Despite the relatively greater importance of boundary effects and fluctuations in two dimensions than in three, the fundamental underlying ideas of particle and continuum mechanics appear to be equally valid in two and three space dimensions. Consider conservation of mass, momentum, and energy. When these three principles are applied locally, to a smooth continuum, the "continuity equation," "equation of motion," and "energy equation" result. Then the phenomenological continuum descriptions of Newtonian viscosity and Fourier heat conduction, coupled with suitable initial *and* boundary conditions, lead to well-posed problems in fluid and solid mechanics.

However, most textbook problems in three-dimensional fluid and solid mechanics are "solved" by using symmetry to reduce them, either analytically or computationally, to more tractable one- or two-dimensional problems. Plane Couette flow is the simplest example. More complex problems involving the unstable growth of interface perturbations—Kelvin-Helmholtz, Rayleigh-Bénard, Rayleigh-Taylor, Richtmeyer-Meshkov, etc. instabilities —are likewise typically solved in two space dimensions. The results are then applied, and compared to real laboratory experiments, in three dimensions. The relationship linking the two-dimensional calculations to three-dimensional experiments would be paradoxical if two-dimensional transport coefficients really diverged. Of course, *fluctuations* in two dimensions are not good representations of their three-dimensional analogs.

In the smoothed-particle approach to numerical solutions of the continuum motion and energy equations, the continuum is replaced by a set of one-, two-, or three-dimensional spatially smoothed particles which obey ordinary differential equations of motion derived from the continuum equations [14]. If two-dimensional particles cannot obey conventional hydrodynamics, the two- and three-dimensional simulations of flows would necessarily disagree with one another. We are in the process of investigating the two- and three-dimensional versions of unsteady Rayleigh-Bénard instability. In the present work we study simpler steady two-dimensional shear flows ("plane Couette flow") in an effort to identify behavior symptomatic of divergence.

III. MODEL

The short-ranged pair potential

$$\phi(r) \equiv 100\varepsilon[1-(r/\sigma)^2]^4 \equiv 100[1-r^2]^4 \, , \quad 0 < r < \sigma \equiv 1$$

has a well-characterized equilibrium fluid-phase equation of state [15], as displayed in Sec. IV, so that it is possible to apply Enskog's hard-disk transport theory [16] to predict nonequilibrium dense-fluid properties. In the present work, we concentrate on a dense-fluid state of unit density—$[N\sigma^2/V \equiv \rho \equiv 1.0; \, \delta\rho \equiv 0.0]$; the particles have unit mass $[m = 1.0]$ and unit reduced energy $[E/N\varepsilon \equiv 1.000; \, \delta\varepsilon \equiv (E/N\varepsilon) - 1.443 = -0.443]$; $(kT/\varepsilon) - 1 \equiv \delta\tau$ is -0.310. See Sec. IV for the complete equilibrium equation of state for the fluid.

We have previously simulated small-system shear flows, for this same force law, with both moving corrugated walls [7] and periodic Lees-Edwards boundary conditions [6,9]. The two approaches showed one qualitative difference. The largest Lyapunov exponent λ_1, which describes the time-averaged rate, $\propto \exp(\lambda_1 t)$, at which two nearby phase-space trajectories separate, appears to diverge logarithmically, $\lambda_1 \propto \ln N$ for N particles, when corrugated boundaries are used [7]. In the periodic case there is much less size dependence. See Fig. 1, which is based on the data appearing in Tables I and II. In this periodic case, it is possible, but far from definitely established, that the largest Lyapunov exponent increases logarithmically. If so, the dependence is very weak, $\lambda_1 \approx 0.001 \ln N$. Our corrugated-boundary results indicated a coefficient orders of magnitude larger. Thus the boundary conditions play a crucial role in determining the system's Lyapunov spectrum.

Certainly the simplest hydrodynamic flow is a steady uniform shear. In the prototypical case the stream-velocity component in the x direction, u_x, varies linearly with y. As the strain rate $\dot{\varepsilon} \equiv du_x/dy$ approaches zero,
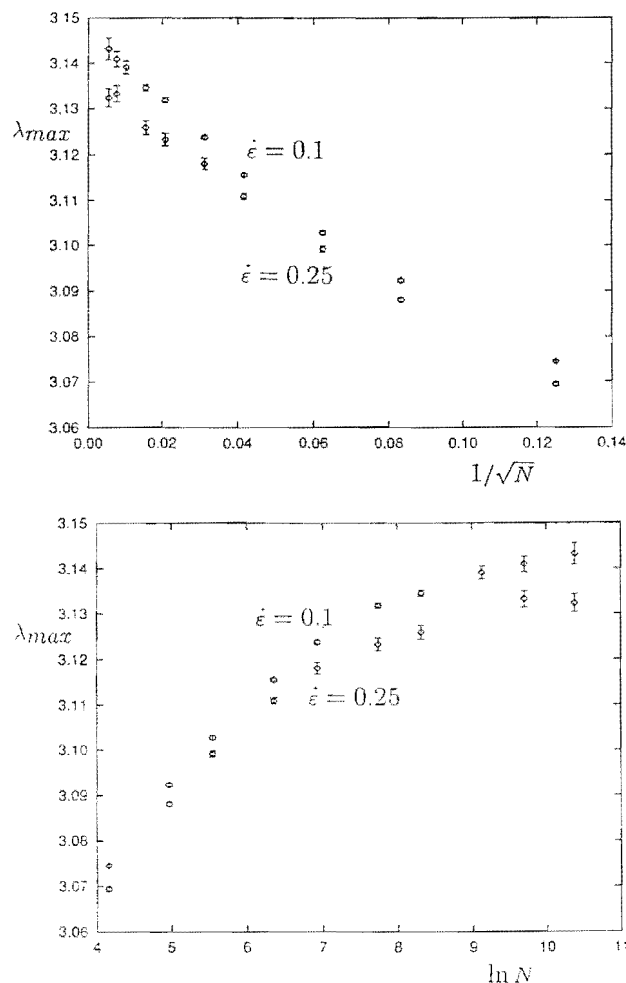

FIG. 1. Dependence of the largest Lyapunov exponent on system size at strain rates of $0.10(\varepsilon/m\sigma^2)^{1/2}$ and $0.25(\varepsilon/m\sigma^2)^{1/2}$. The data shown span the range from $N=64$ to 32 400.

TABLE I. Potential energy, shear viscosities, largest Lyapunov exponent, and shear-stress fluctuations for square periodic two-dimensional systems of N unit mass particles at unit density. The pair potential is $100(1-r^2)^4$. The steady shear strain rate du_x/dy for all of these plane Couette flows is 0.10 and the total energy per particle $E/N \equiv [\Phi+K]/N$ is 1.000 in each case. The mean-squared fluctuation in the shear stress, $\langle \Delta P_{xy}^2 \rangle$, is expected to vary as $1/N$. The maximum shear for each run is indicated, where unit shear requires 2000 time steps of 0.005 each.

N	Φ/N	λ_1	η	$\langle -P_{xy} \rangle$	$\langle N\Delta P_{xy}^2 \rangle$	$\varepsilon_{\max}$
64	0.3052	3.074	1.291	0.129	8.52	40 000
144	0.3042	3.092	1.302	0.130	8.47	18 000
256	0.3039	3.103	1.307	0.131	8.44	11 000
576	0.3036	3.116	1.312	0.131	8.43	10 000
1024	0.3035	3.124	1.312	0.131	8.45	6000
2304	0.3034	3.132	1.319	0.132	8.41	3000
4096	0.3034	3.135	1.315	0.131	8.41	1800
9216	0.3034	3.139	1.319	0.132	8.42	570
16 384	0.3033	3.141	1.317	0.132	8.44	270
32 400	0.3033	3.143	1.319	0.132	8.55	140
65 536	0.3033		1.324	0.132	8.55	200
146 689	0.3033		1.323	0.132	8.41	100
264 196[a]	0.3033		1.322	0.132	8.61	30

[a]This number, 514×514, rather than 512×512, was used to improve the computational efficiency of the simulation.

both Sllod and Dolls algorithms produce equal linear shear viscosities, but the two approaches differ in nonlinear responses, of orders $\dot{\varepsilon}^2$ and $\dot{\varepsilon}^3$, to shear [17] (Evans's [8,17] term *Sllod* refers to its close relationship to the Dolls tensor algorithm). We have used the Sllod approach, with Coriolis's acceleration $-\dot{\varepsilon}p_y$,

$$dx/dt = (p_x/m) + \dot{\varepsilon}y, \quad dy/dt = (p_y/m) ,$$

$$dp_x/dt = F_x - \dot{\varepsilon}p_y - \zeta p_x, \quad dp_y/dt = F_y - \zeta p_y ,$$

$$\zeta \equiv -\dot{\varepsilon}P_{xy}V/2K, \quad K \equiv \sum (p^2/2m) ,$$

$$P_{xy}V \equiv \sum (p_x p_y/m) - \sum (x_{ij}y_{ij}\phi'_{ij}/r_{ij}) ,$$

and the similar alternative Dolls tensor algorithm, in which Coriolis's force contribution to dp_x/dt, $-\dot{\varepsilon}p_y$, is replaced with a similar contribution, $-\dot{\varepsilon}p_x$, to dp_y/dt. In either case, the kinetic part of the pressure-tensor component P_{xy} includes a sum over all N particles, while the potential contribution is a sum over all $N(N-1)/2$ distinct particle pairs. The friction coefficient ζ is chosen so as to maintain the internal energy, $E=K+\Phi$, constant in time. There appears to be no significant difference between the two approaches at a strain rate of $0.10(\varepsilon/m\sigma^2)^{1/2}$. For simplicity, we tabulate only those results based on the Sllod algorithm.

In describing the results of our numerical work we choose units with ε, m, and σ, all equal to unity. (Notice that there is no connection between the potential energy parameter ε and the shear strain rate $\dot{\varepsilon}$.) We use the classic fourth-order Runge-Kutta integrator with a time step of $dt=0.005$. A force with *three* continuous derivatives at the cutoff radius introduces "local" (one integration

step) trajectory errors of order dt^5. These are of the same order as those from the integrator, and so do not degrade trajectory accuracy. The linked-list method [18], in combination with Lees-Edwards's periodic shearing boundary conditions [9], was used.

IV. ENSKOG THEORY

Enskog's theory of dense-fluid transport has survived 70 years' effort to supplant it. The theory proceeds by scaling the low-density results of the Boltzmann equation to high density, including both kinetic and collisional contributions to transport. For a general two-dimensional fluid it is first necessary to use the fluid's thermal pressure, $T(\partial P/\partial T)_V$, to estimate the collisional part of the hard-disk pressure:

$$[PV-NkT]_{\text{disks}} \equiv [TV(\partial P/\partial T)_V - NkT]_{\text{fluid}} .$$

At unit reduced density and temperature $[\rho \equiv N\sigma^2/V \equiv 1; \; kT/\varepsilon \equiv 1]$, $PV/N\varepsilon$ and $E/N\varepsilon$, respectively, are 5.04 and 1.44. In the vicinity of this standard state, the following expansions apply:

$$PV/N\varepsilon = 5 + 8\delta\rho + 2.5\delta\varepsilon + 9\delta\rho^2 + 2\delta\rho\delta\varepsilon ,$$

$$kT/\varepsilon = 1 - \delta\rho + 0.7\delta\varepsilon - 0.8\delta\rho^2 - 0.5\delta\rho\delta\varepsilon ;$$

$$e/\varepsilon \equiv E/N\varepsilon$$
$$= 1.443 + 1.5\delta\rho + 1.5\delta\tau + 2.4\delta\rho^2 + 1.2\delta\rho\delta\tau ,$$

$$\delta\rho \equiv (N\sigma^2/V) - 1.000, \quad \delta\varepsilon \equiv (E/N\varepsilon) - 1.443 ,$$

$$\delta\tau \equiv (kT/\varepsilon) - 1.000 .$$

For the conditions of our simulations, $\delta\rho$ is 0.0 and $\delta\varepsilon$ is -0.443, so that this approximate equation of state predicts that NkT is $[1-0.443\times0.7]N\varepsilon = 0.69N\varepsilon$, in good agreement with the data from our simulations. The corresponding thermal pressure contribution to PV is $2.5\times1.5N\varepsilon = 5.4NkT$.

For numerical evaluations, we use the $V/V_0 = 1.65$ entry from Ree and Hoover's hard-disk fluid table [19], so that our soft potential corresponds to a hard-disk fluid expanded about 65% from close packing. The corresponding reduced compressibility factor of 5.1 has a second virial coefficient contribution of 1.10:

$$[PV/NkT]_{\text{disks}}$$
$$= 1.00 + 1.10 + 0.94 + 0.71 + 0.49 + \cdots = 5.1 .$$

The hard-disk viscosity coefficient, according to Gass [16], can then be written as the sum of three terms:

$$\eta_{\text{disks}} = 0.28(mkT/\sigma^2)^{1/2}(1.10)[0.24+1.00+3.58] ,$$

where the ratio of the kinetic to the potential contribution $(0.24+0.50)/(0.50+3.58)$, is about $1/5$. Using $(kT/\varepsilon)_{\text{disks}} = 0.69$ and $\sigma_{\text{disks}} = 0.78(V/N)^{1/2}$ leads to a numerical viscosity estimate 1.6, which lies about 20% higher than our simulation results.

For the thermodynamic state we have investigated we have also estimated the sound velocity c (used in Sec. V) from our approximate equation of state. The calculation

is most easily carried out by computing the isentropic equation-of-state derivative

$$c^2 = (\partial P / \partial \rho)_S$$

by iterating the [Rankine-Hugoniot] relation

$$\Delta E = E_2 - E_1 = \langle P \rangle \Delta V \equiv (P_2 + P_1)(V_1 - V_2)/2$$

for neighboring values of the volume, V_1 and V_2. The resulting sound velocity is $c = 4.55(\varepsilon/m)^{1/2}$.

V. RESULTS

Keyes's old mode-mode coupling estimate [4] of the logarithmic fluctuation contributions to the viscosity for a similar thermodynamic state was 7% for an increase of just over tenfold in N. In the present work we have accumulated usable viscosity data over a larger range in N, covering more than three orders of magnitude. In Fig. 2 we display the viscosities, with expected errors indicated, as a function of $N^{-1/2}$ and $\ln N$. Based solely on Keyes's old estimate for the dependence on $\ln N$, the expected change in the viscosity η should be approximately 20%

FIG. 2. Steady isoenergetic shear viscosities for N two-dimensional particles interacting with the potential $100\varepsilon[1 - (\sigma/r)^2]^4$ with an internal energy equal to $N\varepsilon$. The error bars indicate estimated standard deviations for the viscosities. The number of particles ranges from 64 to 264 196.

in the range of $\ln N$ shown in the figure. In the region where the data vary smoothly, from about 1024 to 264 196 particles, a change of 0.03 $\ln N$ would lead to an increase exceeding 10%. Instead, our data show a much smaller increase for these larger values of N.

Our numerical results are given in Tables I and II. The shear viscosity coefficients $\{\eta(N)\}$ for $N = 64, 144, 256, 576, \ldots, 264\,196$ were all determined with expected errors substantially less than 1% as shown in Fig. 2. We have suppressed all of our viscosity results for even smaller systems. The irregular nature of the data for the smaller systems together with the lack of significant N dependence for the larger systems rule out a meaningful least-squares fit, though the data do suggest, as shown at the top of Fig. 2, an inverse dependence on system size, $\propto N^{-1/2}$. It is clear from our data that there is no logarithmic dependence of viscosity on system size, even at a level one or two orders of magnitude below the old mode-coupling predictions. There is instead a very modest shear rate dependence, hardly large enough to be worth including in hydrodynamic simulations.

We conclude that the shear viscosity is well defined, in two dimensions, when a global constraint of constant energy is used to stabilize the hydrodynamic state. Neither N dependence nor hydrodynamic instabilities are observed, at Reynolds numbers as high as 50 000. As we emphasized earlier [6], there is numerical evidence that our periodic boundary condition is much less intrusive than is a rigid one. Here we find no evidence whatsoever for the logarithmic transport divergence predicted by the kinetic and mode-coupling theories [1]. It is therefore very desirable that those theories be extended to take into account global boundary conditions of the type used here.
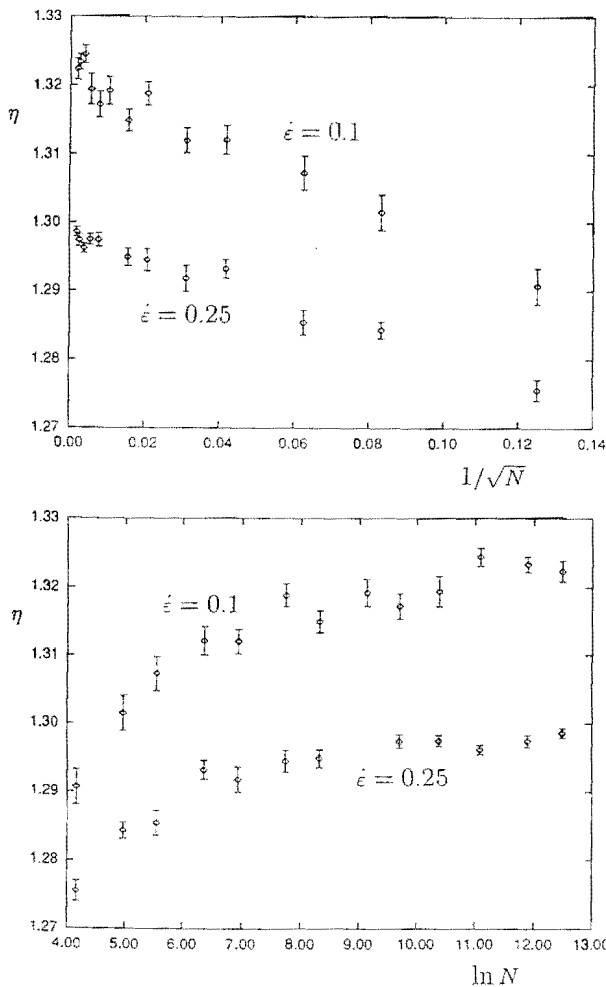
TABLE II. Potential energy, shear viscosities, largest Lyapunov exponent, and shear-stress fluctuations for square periodic two-dimensional systems of N unit mass particles at unit density. The pair potential is $100(1 - r^2)^4$. The steady shear strain rate du_x/dy for all of these plane Couette flows is 0.25, and the total energy per particle $E/N \equiv [\Phi + K]/N$ is 1.000 in each case. The mean-squared fluctuation in the shear stress, $\langle \Delta P_{xy}^2 \rangle$, is expected to vary as $1/N$. The maximum shear for each run is indicated, where unit shear requires 800 time steps of 0.005 each. Trials with $N = 4096$ and steps of 0.01 produced results not significantly different from these.

N	Φ/N	λ_1	η	$\langle -P_{xy} \rangle$	$\langle N\Delta P_{xy}^2 \rangle$	ε_{max}
64	0.3073	3.069	1.276	0.319	8.65	50 000
144	0.3064	3.088	1.284	0.321	8.61	30 000
256	0.3062	3.099	1.285	0.321	8.58	10 000
576	0.3060	3.111	1.293	0.323	8.57	6200
1024	0.3059	3.118	1.292	0.323	8.61	4000
2304	0.3058	3.123	1.295	0.324	8.56	1200
4096	0.3058	3.126	1.295	0.324	8.52	1000
16 384	0.3058	3.133	1.297	0.324	8.64	530
32 400	0.3058	3.132	1.297	0.324	8.72	330
65 536	0.3058		1.296	0.324	8.53	250
146 689	0.3058		1.297	0.324	8.52	60
264 196	0.3058		1.299	0.325	8.26	50

VI. DISCUSSION

The long-standing view that two-dimensional transport coefficients *diverge*, based on analyzing the Green-Kubo expressions [20] for those coefficients, does not take the boundaries into account, despite the fact that both boundaries and fluctuations make contributions of order $N^{-1/2}$ to the intensive properties of two-dimensional N-body systems. Our work here treats systems with global boundary conditions. Our viscosity results suggest that the predicted divergence, *if* it exists in any case at all, must be boundary dependent.

It is of course possible that a real long-time divergence could be masked in our own computer simulations. The simulations include three characteristic times, the "deformation time" $1/\dot{\epsilon}$, the "sound traversal time" $V^{1/2}/c$, and the "diffusive traversal time" $V(\rho/\eta)$. For our largest simulation and at the lowest strain rate these times are roughly 10, 113, and 200 000. If stress-stress correlations were broken at the deformation time, one might expect to find a significant increase in viscosity at lower rates, but still with no significant number dependence at any fixed rate.

After most of our numerical work had been completed, we became aware of related work on the Green-Kubo and subtraction-technique evaluation of two-dimensional shear viscosities [12]. Gravina, Ciccotti, and Holian conclude, from their studies on relatively small systems, that there is no evidence for a substantial rate dependence either. Where the error can be in the simple diffusion-equation arguments predicting a logarithmic divergence remains a mystery.

There is certainly the possibility that Green-Kubo and nonequilibrium simulations measure different transport coefficients, so that one method or the other may show more number dependence or larger fluctuations. The work presented here, together with the complementary work of Gravina, Ciccotti, and Holian, suggests that all of the current computational approaches agree that viscosity is a meaningful material property in two dimensions. Real laboratory experiments, on thin films, could provide a welcome confirmation of these results.

There is a different divergence, which is real, significant, and easily quantified, associated with the nonequilibrium phase-space probability density itself, in both two- and three-dimensional systems. Consider shearing a system, initially at equilibrium, in the absence of ergostats or thermostats:

$$dx/dt = (p_x/m) + \dot{\epsilon}y, \quad dy/dt = (p_y/m),$$

$$dp_x/dt = F_x - \dot{\epsilon}p_y, \quad dp_y/dt = F_y.$$

In this case, the thermodynamic work performed by the two terms linear in $\dot{\epsilon}$, during a time interval Δt, is $\Delta E = -\int_0^{\Delta t} P_{xy} V \dot{\epsilon}\, dt$, so that the energy at that time exceeds the initial equilibrium energy E_0. Because the comoving probability $f d\Gamma$, initially proportional to $\exp(-E_0/kT)$, is not changed by the adiabatic work, this probability can be expressed in terms of the equilibrium probability for the energy at $t = \Delta t$. In terms of the current energy, $E_0 + \Delta E$, we have

$$f_{\text{nonequilibrium}}/f_{\text{equilibrium}} = \exp(+\Delta E/kT).$$

In the case that an ergostat is used to maintain the energy constant, the probability density, relative to the equilibrium one, also changes, and by exactly the same amount. This result follows directly from the integration of Liouville's theorem in D dimensions:

$$d\ln f/dt = -\sum [\partial\dot{q}/\partial q + \partial\dot{p}/\partial p]$$
$$= -\sum \partial\dot{p}/\partial p = ND\zeta$$
$$\rightarrow f_t/f_0 = \exp[-\dot{\epsilon}(NDV)$$
$$\times \int_0^t (P_{xy}/2K)dt'].$$

Then the ensemble-averaged energy change, from integrating $(fP_{xy})_{t'}$ up to time t is given, in the linear approximation, by the autocorrelation integral of the rate-of-energy change. This leads directly to the macroscopic Green-Kubo identification of the shear viscosity:

$$\eta(t\rightarrow\infty) = (V/kT)$$
$$\times \int_0^{t\rightarrow\infty} \langle P_{xy}(0)P_{xy}(t')\rangle_{\text{equilibrium}} dt',$$

as well as to the conclusion that the phase-space distribution is divergent on a multifractal phase-space attractor [6,21]. For our isoenergetic simulations, it is straightforward to show that the comoving nonequilibrium phase-space probability density f continually increases with time:

$$d\ln f/dt \equiv -\dot{\epsilon}P_{xy}V/kT.$$

In the long-time limit the time-averaged right hand side approaches the intrinsically positive quantity $\eta\dot{\epsilon}^2 V/kT$, showing that $\ln f$ diverges, exponentially fast, on the average, with time. This continuing increase of $\ln f$ with time signals the divergence of f as it collapses onto a multifractal strange attractor [6]. Perhaps this collapse can somehow help to limit the size of fluctuations. The number dependence of the divergence of f has recently been well documented [6].

Because the boundaries are not considered in the derivation, the long-time-tail divergence (as opposed to the phase-space density divergence) might be suspect. Here we have seen that the numerical shear viscosities for two-dimensional systems of between 32^2 and 514^2 particles provide no evidence for such number dependence.

We conclude that the logarithmic divergence of two-dimensional transport coefficients is implausible, both on physical and numerical grounds. This finding, after some 25 years of nonequilibrium simulations, fully supports Barker's assessment [22] of the relationship between Green-Kubo and nonequilibrium simulations:

"It seems to me that to constrain the molecular dynamicist to use only Green-Kubo is somewhat analogous to constraining the experimentalist to measure the viscosity by studying only the decay of spontaneously arising velocity fluctuations. If this requires a theory of its own, so be it!" We hope that these results will help to stimulate the theoretical advances recommended by Barker. In the meantime there would seem to be no

problem in applying two-dimensional transport coefficients to the simulation of two-dimensional hydro-dynamic flows.

Note added in proof. Onuki [23] and Ernst, Cichocki, Dorfman, Sharma, and van Beijeren [24] have independently derived finite two-dimensional viscosities which contain a logarithmic dependence on strain rate consistent with our results.

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