Shear-rate dependence of the viscosity for dilute gases

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The non-Newtonian viscosity of a dilute gas of hard spheres under uniform shear flow is obtained by means of a Boltzmann–Monte Carlo simulation method. The influence of initial conditions is analyzed. The results are compared with exact solutions of the Boltzmann and the Bhatnagar-Gross-Krook equations. The agreement is shown to be fairly good over a wide range of shear rates.

I. INTRODUCTION

Nonlinear transport beyond the scope of Navier-Stokes equations is a very interesting problem from both a theoretical and an experimental point of view, particularly when dealing with molecular or rheological fluids.[1] On the other hand, for simple atomic fluids, nonlinear effects have been observed in computer simulations for the case of uniform shear flow. The corresponding macroscopic state has a constant linear-velocity profile with spatially uniform temperature and density, while the temperature monotonically increases in time due to viscous heating. This state is generated at the microscopic level by imposing Lees-Edwards boundary conditions,[2] that are, in fact, equivalent to periodic boundary conditions in the local Lagrangian frame. Furthermore, the dynamics of the particles is governed by classical-mechanics equations.[3,4] Since the shear rate can take arbitrarily large values in this state, deviations from Newton's viscosity law can be observed.

The shear-rate dependence of the shear viscosity in the above state has been derived by Ikenberry and Truesdell for a system of Maxwell molecules described by the Boltzmann equation.[5] For more general interaction potentials, Zwanzig has obtained a closed differential equation for the shear viscosity by using the Bhatnagar-Gross-Krook (BGK) model kinetic equation.[6] These are the only exact kinetic theory calculations for uniform shear flow of which we are aware. On the other hand, this state has been extensively studied by means of molecular-dynamics methods.[7,8] Most of these simulations deal with dense fluids, although some efforts have been carried out recently for dilute gases.[9]

In this paper we report the results for the shear viscosity generated by a Boltzmann–Monte Carlo simulation method[10] for a system of hard spheres. This method is especially devised to mimic the Boltzmann equation. A clear description can be found in Ref. 10. To the best of our knowledge, this method has not been applied up to now to the uniform-shear-flow state. It must be noticed that, in contrast to most of previous works,[7–9] we do not introduce any kind of artificial thermostat to prevent viscous heating. The price to be paid is that the system does not reach a stationary state.

Two main points are considered in this study. First, we will address the question of the existence of the non-Newtonian shear viscosity function, independent of the initial conditions and characterized only by the interatomic force law. Since the uniform shear flow is a time-dependent state, it is not clear *a priori* whether the influence of the initial conditions is restricted to a "short" initial layer $(t < t_h)$, after which the system reaches a hydrodynamic stage. This point has not been explicitly considered in previous simulations; usually the system is just aged for a more or less arbitrarily large enough period of time. We will see that it is possible to identify a hydrodynamic shear viscosity over a rather wide range of values of a reduced shear rate. This viscosity function can be expected to be the one corresponding to the so-called "normal" solution of the Boltzmann equation for hard spheres. The usual way of constructing the normal solution, by means of the Chapman-Enskog expansion, is only useful for small shear rates.[11] Then, our second point is to compare the simulation data with the known exact solutions mentioned above: the Boltzmann solution for Maxwell molecules (BM solution[5]) and the solution of the BGK model kinetic equation for a system of hard spheres (BGK-HS solution).[6,11] The results show that both models lead in fact to quite a good agreement with the simulation data.

II. UNIFORM SHEAR FLOW

The conservation laws for a dilute gas under uniform shear flow lead to an increase of temperature given by

$$\frac{dT(t)}{dt} = -\frac{2}{3nk_B}aP_{xy}(t), \qquad (1)$$

where T is the temperature, n is the density, k_B is Boltzmann's constant, a is the shear rate and P_{xy} is the xy component of the pressure tensor. A coordinate system with the x axis along the direction of the flow has been chosen. The heating equation (1) shows that the state of the gas under uniform shear flow depends on time due to viscosity. We can define a generalized shear viscosity coefficient η for all times by

$$\eta(t) = -P_{xy}(t)/a \qquad (2)$$

However, at sufficiently large times $\eta(t)$ is expected to depend on time just through $T(t)$: $\eta(t) = \eta(a, T(t))$. This

characterizes the time domain where $\eta(t)$, as defined by Eq. (2), becomes a hydrodynamic shear viscosity. Then, dimensional analysis for hard spheres leads to the form

$$\frac{\eta(a,T(t))}{\eta_{NS}(T(t))}=\eta^*(a^*), \quad a^*(T(t))=\frac{a}{v(T(t))}, \quad (3)$$

where η^* is a dimensionless function of the reduced shear rate a^*. In Eq. (3), $\eta_{NS}(T)=\frac{5}{16}(mk_B T/\pi)^{1/2}/\sigma^2$ is the Navier-Stokes shear viscosity from the Boltzmann equation for hard spheres of mass m and diameter σ, and $v(t)=nk_B T/\eta_{NS}(T)$ is an effective collision frequency. Notice that both reduced variables, η^* and a^*, change with time. In particular, Eqs. (1) and (3) show that a^* monotonically decreases in time.

In principle, if one uses the values of $P_{xy}(t)$ and $T(t)$ to compute η^* as a function of a^*, one will have a different function for each trajectory of the system, i.e., for each initial condition. However, if the system reaches a hydrodynamic regime, all the particular functions $\eta^*(a^*)$ must tend to coincide for values of a^* sufficiently smaller than the initial $a_0^* \equiv a^*(T(0))$. As stated in Sec. I, the check of this expectation is one of the main goals of this paper.

III. THE SIMULATION

In our simulation we have considered a system containing 2000 particles between two plates separated by a distance of 10λ, where $\lambda=(\sqrt{2}\pi n\sigma^2)^{-1}$ is the mean free path for hard spheres. In the Boltzmann–Monte Carlo simulation method,[10] it is necessary to split the system into cells and also to use a time step. Because of the symmetry of the problem, the cells can be taken as layers parallel to the plates. In most of the simulations we have taken a layer width of $\frac{1}{4}\lambda$. On the other hand, the fact that the collision frequency increases in time suggests the use of a nonconstant time step. It is then convenient to introduce a new time scale defined by

$$s=\int_0^t dt' v(T(t')). \quad (4)$$

Thus $s(t)$ represents the average number of collisions per particle between 0 and t. In this variable, we have considered a constant step Δs ($\Delta s=0.016/\sqrt{\pi}$ in most of the simulations). That means that the corresponding real time interval Δt behaves as $[T(t)]^{-1/2}$. Finally, the Lees-Edwards boundary conditions have been employed to produce uniform shear flow. To improve the statistics, a number of different trajectories have been generated for each initial state. The reported results are typically averaged over 70 trajectories. In all cases, the macroscopic hydrodynamic profiles defining the uniform shear flow were obtained within typical deviations of the order of 1% in each layer.

The shear viscosity is computed from P_{xy} using Eq. (2). Two different methods have been considered to evaluate P_{xy}: first, directly from its definition as the momentum flux; second, from the rate of change of the temperature by means of Eq. (1). The agreement between both methods has been employed as a test of the accuracy of the simulation. Figure 1 shows the results obtained for
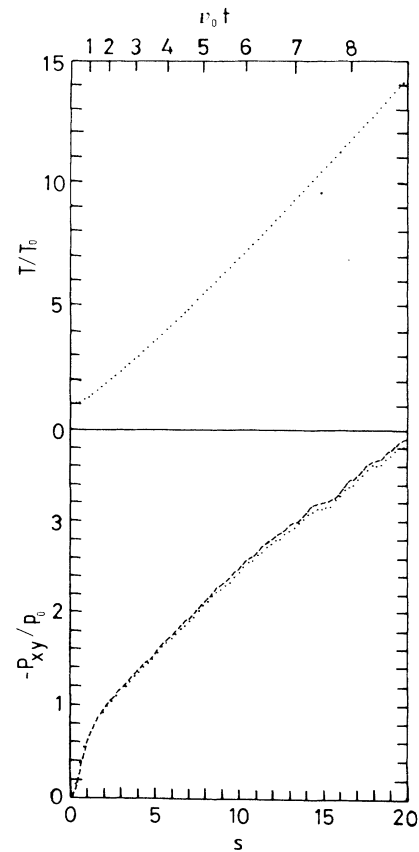

FIG. 1. Time evolution of the temperature T and the pressure tensor element P_{xy} evaluated directly as the momentum flux (dots) and from the heating equation (broken line). The initial condition is of local equilibrium with $a_0^*=5\sqrt{\pi}/8$. In the graph, T_0, $P_0=nk_B T_0$, and v_0 stand for the initial values of the temperature, the pressure, and the collision frequency, respectively.

an initial velocity distribution of local equilibrium with $a_0^*=5\sqrt{\pi}/8$. We find good agreement between the two methods; their relative discrepancy is always less than 5%. We have studied the influence on the result of the values chosen for the width of the layer and the time step, finding that the method based on Eq. (1) is less sensitive to these values. Therefore, in the following, only results based on Eq. (1) will be reported.

IV. RESULTS AND DISCUSSION

From $T(t)$ and $P_{xy}(t)$ one can compute a^* and η^* at any time and then plot η^* versus a^*. Figure 2 compares the results for initial distributions of local equilibrium with $a_0^*=5\sqrt{\pi}/8$, $5\sqrt{2\pi}/8$, and $5\sqrt{\pi}/4$. Of course, each curve starts at $a^*=a_0^*$ and cannot be extrapolated to larger values of a^*. As said before, we want to identify the hydrodynamic region, i.e., the part of the curves in Fig. 2 that does not depend on the particular initial condition. We see that the three curves overlap for $a^* \lesssim 0.5$,
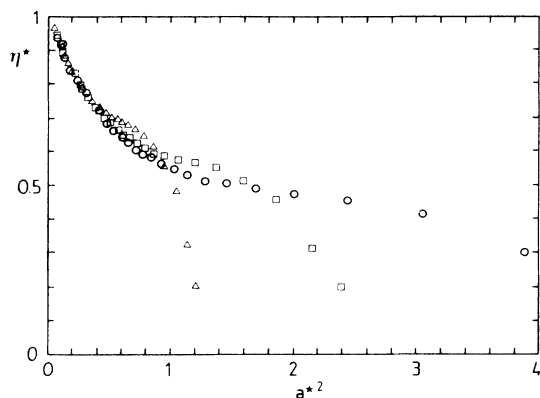
FIG. 2. Simulation data of the reduced shear viscosity versus the square of the reduced shear rate for initial conditions of local equilibrium with $a_0^* = 5\sqrt{\pi}/8$ ($\triangle$), $5\sqrt{2\pi}/8$ ($\square$), and $5\sqrt{\pi}/4$ ($\bigcirc$).
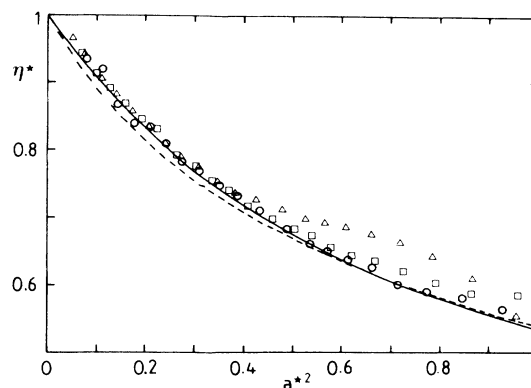
FIG. 3. Same as Fig. 2. The theoretical curves corresponding to the Boltzmann equation for Maxwell molecules (broken line) and to the BGK equation for hard spheres (solid line) are also plotted.

and therefore, the values of η^* in that region can be considered as hydrodynamic. Of course, one expects that the hydrodynamic part of a particular curve can be enlarged if one starts from a larger initial value a_0^*. Therefore, the representative part of the curve corresponding to $a_0^* = 5\sqrt{\pi}/4$ is expected to extend somewhat to values of a^* larger than 0.5. In any case, it seems clear that the system needs a certain period of time to reach the hydrodynamic regime. We have estimated from our data that this time is of the order of four average collision times ($s \simeq 4$). For longer times, the non-Newtonian shear viscosity is then expected to correspond to the normal solution of the Boltzmann equation. Also, Fig. 2 shows that η^* tends to 1 as a^* goes to zero, in agreement with theory. Notice that the limit $a^* \to 0$ is not accessible in the simulation since it corresponds to $s \to \infty$.

The region $a^* < 1$ is amplified in Fig. 3. Also, the BGK-HS solution[11] and the BM solution[5] are plotted. In both cases, a^* and η^* are still defined by Eq. (3) but using the corresponding expression for $\eta_{NS}(T)$. For $a^* \lesssim 0.5$, both models fairly reproduce the simulation data, the fit being slightly better for the BGK hard-sphere model. The agreement extends up to $a^* = 1$ if the simulation corresponding to $a_0^* = 5\sqrt{\pi}/4$ is considered. Both exact solutions exhibit the behavior $\eta^* \sim a^{*-4/3}$ for asymptoti-

cally large a^*.[5,11] Whether this also holds in the Boltzmann equation for hard spheres cannot be easily inferred from the simulation method since it would require one to start from an extremely large initial value a_0^* in order to be still in the region of large a^* after an aging period of $s \simeq 4$. Several conclusions follow from the above results. The comparison with the BM solution shows that the shear viscosity obtained from the Boltzmann equation seems to be rather insensitive to the interaction potential over a wide range of shear rates when appropriate reduced units are used. Moreover, the BGK equation is a fairly good approximation of the Boltzmann equation for this problem. These conclusions are reinforced by the fact that, for Maxwell molecules, the shear viscosity obtained from the Boltzmann equation is the same as the one obtained from the BGK model.[6] Work is now in progress to study whether this agreement also holds when the complete description, given by the velocity distribution function, is considered.

ACKNOWLEDGMENTS

[1] R. Bird, R. Armstrong, and O. Hassager, Dynamics of Polymeric Liquids (Wiley, New York, 1977).

[2] A. Lees and S. Edwards, J. Phys. C 5, 1921 (1972).

[3] J. W. Dufty, A. Santos, J. J. Brey, and R. F. Rodriguez, Phys. Rev. A 33, 459 (1986).

[4] J. W. Dufty, J. J. Brey, and A. Santos, in Molecular-Dynamics Simulation of Statistical-Mechanical Systems, edited by G. Ciccotti and W. G. Hoover (North-Holland, Amsterdam, 1986), pp. 294–303.

[5] E. Ikenberry and G. Truesdell, J. Rat. Mech. Anal. 5, 55 (1956); G. Truesdell and R. G. Muncaster, Fundamentals of Maxwell's Kinetic Theory of a Simple Monatomic Gas

(Academic, New York, 1980).

[6] R. Zwanzig, J. Chem. Phys. 71, 4416 (1979).

[7] T. Naitoh and S. Ono, J. Chem. Phys. 70, 4515 (1979); J. Erpenbeck, Physica 118A, 144 (1983); W. G. Hoover, Annu. Rev. Phys. Chem. 34, 103 (1983).

[8] J. P. Ryckaert, A. Bellemans, G. Ciccotti, and G. V. Paolini, Phys. Rev. Lett. 60, 128 (1988).

[9] W. Loose and S. Hess, Phys. Rev. Lett. 58, 2443 (1987); Phys. Rev. A 37, 2099 (1988).

[10] G. Bird, Molecular Gas Dynamics (Clarendon, Oxford, 1976).

[11] A. Santos, J. J. Brey, and J. W. Dufty, Phys. Rev. Lett. 56, 1571 (1986).