Non-Boltzmann behavior from the Boltzmann equation

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We compute the stress autocorrelation function in a two- and three-dimensional system by using the lattice-Boltzmann method. The algebraic long-time behavior $\sim t^{-d/2}$ in the stress correlation function is clearly observed. The amplitude of this tail is compared with the mode-coupling expression for the long-time tail in the stress correlation function. Agreement is found between the mode-coupling theory and simulation in both two and three dimensions.

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I. INTRODUCTION

In 1970 Alder and Wainwright [1] reported the results of a computer simulation study of the decay of velocity fluctuations in a hard-sphere fluid. These simulations revealed that velocity fluctuations do not decay exponentially, as had been previously assumed, but algebraically. This observation was of great importance because nonexponential decay of the velocity autocorrelation function (velocity ACF) is not compatible with Boltzmann's "molecular chaos" hypothesis, i.e., the assumption that there is no correlation between the velocity of a particle at time t and the velocity of its collision partners at any later time.

Subsequently, mode coupling [2] and kinetic theories [3] were developed to provide a theoretical framework for the description of long-time tails in correlation functions. Both classes of theory reproduce the algebraic decay of the velocity, ACF, $\phi(t)\sim t^{-d/2}$, where d is the dimensionality of the fluid and t the time. In addition, the same theories also predict an algebraic long-time tail in the stress correlation function. The mode-coupling theory prediction for the asymptotic form of the stress autocorrelation function is [2]

$$\phi_{xy}(t)=\frac{1}{\rho d(d+2)}\left[\frac{d^2-2}{(8\pi\nu t)^{d/2}}+\frac{1}{(4\pi\Gamma t)^{d/2}}\right]\equiv\frac{d_0}{t^{d/2}}\ . \tag{1}$$

In this equation, $\phi_{xy}(t)$ is the correlation function for the xy component of the stress tensor, ρ is the number density, ν is the "bare" kinematic viscosity, and Γ is the sound wave damping coefficient. Unlike the long-time tail in the velocity ACF, the algebraic tail in the stress correlation function has not been observed directly either in simulations or in experiment, except in a very simple one-dimensional model [4] that does not really correspond to a fluid.

The most accurate tests of mode-coupling theory in a realistic system were obtained in simulations of a simplified model for an atomic fluid, namely a lattice-gas cellular automaton of the type introduced by Frisch, Hasslacher, and Pomeau [5]. Kadanoff, McNamara, and Zanetti [6] investigated the stress-stress correlation function indirectly by comparing the apparent viscosity of such a fluid with theoretical predictions based on mode-coupling theory. They investigated the system size dependence of the kinematic viscosity, and found the expected logarithmic divergence. For the velocity autocorrelation function, Frenkel and Ernst [7] exploited some special features of the lattice gas and computed the velocity ACF of a tagged particle with an accuracy that was at least four orders of magnitude better than was hiterto possible. Unfortunately, the same approach cannot be used to improve the accuracy of the calculation of the stress autocorrelation function. For the velocity ACF of a tagged particle in a lattice gas, it proved possible to perform an average over *all* possible labelings of the tagged particle. In contrast, no such averaging can be performed in the case of stress, which is a collective, rather than single-particle property. As a consequence, the stress correlation function is very noisy. It would seem attractive to try to improve the statistics of the stress ACF by performing some kind of pre-averaging that does reduce the statistical fluctuations but not the way in which stress decays in the lattice-gas fluid. A natural pre-averaged version of a lattice-gas cellular automaton fluid is the so-called lattice-Boltzmann model [8,9]. The advantage of the lattice-Boltzmann model is that one can study the decay of an initial perturbation of the stress without any statistical noise. The disadvantage is that, due to the pre-averaging, it is no longer a truly atomistic model. Moreover, the pre-averaging has ended all spontaneous fluctuations. Hence, the way to study the stress ACF is not to watch the decay of spontaneous fluctuations in the stress (there are none), but to make use of Onsager's regression hypothesis and study the decay of an imposed perturbation of the stress. In this paper, we report calculations of the stress ACF, using a lattice-Boltzmann model.

At first sight, it may seem strange to look for long-time tails in a Boltzmann model. After all, in the Boltzmann equation that determines the time evolution of this lattice model, one ignores the correlations between successive collisions that, in the kinetic theory description, give rise to long-time tails. Yet, the lattice-Boltzmann model does reproduce the hydrodynamic behavior of a fluid. In the

mode-coupling theories of long-time tails in simple fluids, it is precisely the slow decay of hydrodynamic modes that is responsible for the appearance of long-time tails (that are, for this reason, often referred to as hydrodynamic long-time tails).

II. LATTICE-BOLTZMANN MODEL

The lattice-Boltzmann model is a pre-averaged version of a lattice-gas cellular automaton (LGCA) model of a fluid. In lattice-gas cellular automaton the state of the fluid at any (discrete) time is specified by the number of particles at every lattice site and their velocity. Particles can only move in a limited number of directions (towards neighboring lattice points) and there can be at most one particle moving on a given "link." The time evolution of the LGCA consists of two steps. (1) Propagation: every particle moves in one time step, along its link to the next lattice site. (2) Collision: at every lattice site particles can change their velocities by collision, subject to the condition that these collisions conserve the number of particles and momentum (and retain the full symmetry of the lattice). In the lattice-Boltzmann method (see, e.g., [10]), the state of the fluid system is no longer characterized by the number of particles that move in direction $\mathbf{c}_i$ on lattice site $\mathbf{r}$, but by the *probability* to find such a particle. The single-particle distribution function $n_i(\mathbf{r},t)$ describes the average number of particles at a particular node of the lattice $\mathbf{r}$, at a time t, with the discrete velocity $\mathbf{c}_i$. The hydrodynamic fields, mass density ρ, momentum density $\mathbf{j}$, and the momentum flux density $\mathbf{\Pi}$ are simply moments of this velocity distribution,

$$\rho=\sum_i n_i \ , \quad \mathbf{j}=\sum_i n_i\mathbf{c}_i \ , \quad \mathbf{\Pi}=\sum_i n_i\mathbf{c}_i\mathbf{c}_i \ . \tag{2}$$

The lattice model used in this work is the four-dimensional face-centered hypercubic (FCHC) lattice. A two- or three-dimensional model can then be obtained by projection in the required number of dimensions. This FCHC model is used because three-dimensional cubic lattices do not have a high enough symmetry to ensure that the hydrodynamic transport coefficients are isotropic.

The time evolution of the distribution functions n_i is described by the discretized analogue of the Boltzmann equation [11],

$$n_i(\mathbf{r}+\mathbf{c}_i,t+1)=n_i(\mathbf{r},t)+\Delta_i(\mathbf{r},t) \ , \tag{3}$$

where Δ_i is the change in n_i due to instantaneous molecular collisions at the lattice nodes. The post-collision distribution $n_i+\Delta_i$ is propagated in the direction of the velocity vector $\mathbf{c}_i$. A complete description of the collision process is given in [12]. The main effect of the collision operator $\Delta_i(\mathbf{r},t)$ is to relax the nonequilibrium part of the momentum flux. The nonlinear expression for the local equilibrium momentum flux density $\mathbf{\Pi}^{\rm eq}$ is given by

$$\mathbf{\Pi}^{\rm eq}=p\mathbf{I}+\rho\mathbf{u}\mathbf{u} \ , \tag{4}$$

with p the local pressure, $\mathbf{I}$ the unit tensor, and $\mathbf{u}$ the local fluid velocity. In the linearized version of the equilibrium part of the momentum flux density is given by

$$\mathbf{\Pi}^{\rm eq}=p\mathbf{I} \ . \tag{5}$$

The rate of stress relaxation, or equivalently, the kinematic viscosity v, can be chosen almost freely (as discussed in [9]). In the linear lattice-Boltzmann model [Eq. (5)] Π_{xy} can only decay exponentially. To observe the long-time behavior of the stress ACF, a coupling to the momentum is essential. The second term in Eq. (4), which is usually only taken into account to study high Reynolds number flow, does exactly this. In order to observe the long-time tail in the stress ACF, the full nonlinear stress tensor had to be used in the simulation.

As the lattice-Boltzmann model is purely dissipative, microscopic fluctuations in the fluid are not included. Such fluctuations can be incorporated in the lattice-Boltzmann model by adding a suitable random noise term to the stress [13]. However, for the present work, such fluctuations are not essential for the phenomenon under study yet would seriously deteriorate the statistical accuracy of our calculations.

The stress in the system, which is a collective property, is given by

$$\mathbf{\Sigma}=\sum_{\mathbf{r}}(\mathbf{\Pi}-p\mathbf{I}) \ . \tag{6}$$

For the sake of convenience, we consider only one component of the traceless symmetric part of the stress tensor *viz.* the xy component. Other components give rise to the same correlation functions. We compute the stress ACF by correlating the initial perturbation of the stress with the stress at some later time t,

$$\phi_{xy}(t)=\frac{\langle\Delta\Sigma_{xy}(0)\Delta\Sigma_{xy}(t)\rangle}{\langle[\Delta\Sigma_{xy}(0)]^2\rangle} \ , \tag{7}$$

where $\Delta\Sigma_{xy}=\Sigma_{xy}(t)-\Sigma_{xy}(\infty)$. It is important to subtract the steady state ($t=\infty$) value of the stress tensor because, in a finite system, the initial stress perturbation will relax to a uniform velocity field with an associated stress given by Eq. (4),

$$\Sigma_{xy}(\infty)=\frac{\Sigma_{xy}(0)}{V} \ , \tag{8}$$

where V is the volume of the fluid. In a LGCA, where the stress is purely kinetic in origin, the stress at site $\mathbf{r}$ at time t is uncorrelated to the stress at that same time at any other lattice point. In our calculations we have therefore chosen to consider the simplest possible initial condition *viz.* a small perturbation of the stress *at one lattice site only.*

III. RESULTS

Having set up the system with an initial local stress perturbation, we followed the time evolution of the total stress of the system using the dynamics of the lattice-Boltzmann model. In fact, we did the simulations both for the linearized and the nonlinear expression for the stress tensor. All quantities were measured in lattice units, such that the lattice spacing, time step, and particle

mass are all set to unity.

In order to be able to compare the tail amplitude as obtained from the simulations with the theoretical expression [Eq. (1)] we need to know the sound damping coefficient Γ. At the Boltzmann level, this quantity is given by $\Gamma=2(d-)\nu/d+\zeta$ with ζ being the kinematic bulk viscosity. ν and ξ were "measured" by setting up a sound wave in the system and measuring the decay of that wave in the long wavelength limit [14]. Γ, ζ, and ν were computed for a range of imposed kinematic viscosities between 0.01 and 0.50.

The simulations in two dimensions were performed on a system of 250×250 lattice sites. For this size of simulation box we followed the stress ACF for 140 time steps. This upper limit was chosen because, after this time, interference occurred due to sound waves that cross the periodic system. In Fig. 1, we show the stress ACF of the lattice-Boltzmann model for several different values of the kinematic viscosity and the nonlinear expression for the stress. For this model, we do indeed observe a clear algebraic decay of the stress ACF. As expected for a two-dimensional fluid, the exponent of the algebraic long-time tail was -1. In contrast, *no* algebraic tail is observed if the nonlinear terms in the stress are ignored. This is understandable because in the linearized model there is no mechanism by which the different modes can couple.

The limiting value d_0 was determined by plotting $t\phi_{xy}(t)$ as a function of $1/t$. The intercept for $1/t=0$ yields the desired amplitude. The results of this analysis are shown in Fig. 2. In this figure, we also show the theoretical tail coefficient given by Eq. (1). Figure 2 shows that the mode-coupling predictions of the tail coefficient are in excellent agreement with the simulation results. The discrepancy between the mode-coupling theory and the simulation results is less than one tenth of a percent.

The simulations in three dimensions were performed on a system of $90\times90\times90$ lattice sites. For this size simulation box we followed the stress ACF for times up to $t=50$. After this time, interference due to the roundtrip of sound waves occurred. In the three-dimensional fluid, we also find algebraic decay of the stress ACF in the nonlinear lattice-Boltzmann model only. The algebraic tail is characterized by an exponent -1.5, as expected. Figure 3 shows the stress ACF of the three-dimensional lattice-Boltzmann fluid for several values of the kinematic viscosity.

We performed the same extrapolation procedure as described above to determine the amplitude of the long-time tail. Specifically, we plotted $t^{3/2}\phi_{xy}(t)$ as a function of $1/t$. As before, the amplitude of the algebraic tail d_0 is

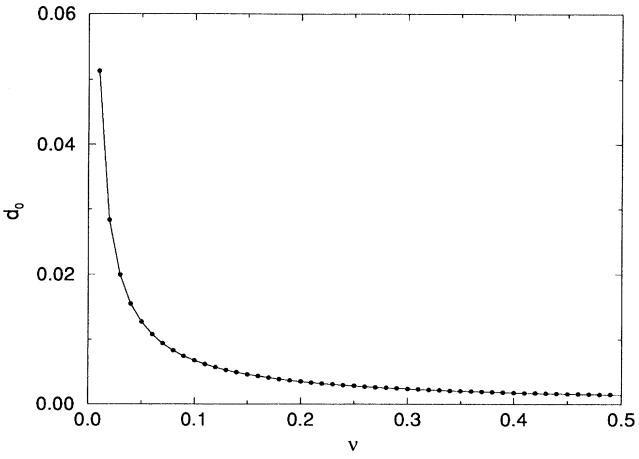

FIG. 2. The tail coefficient of the stress ACF for a two-dimensional lattice-gas fluid, as a function of the dimensionless kinematic viscosity ν. The points are the results of simulations of the lattice-Boltzmann model, while the drawn curve corresponds to the prediction of the mode-coupling theory.

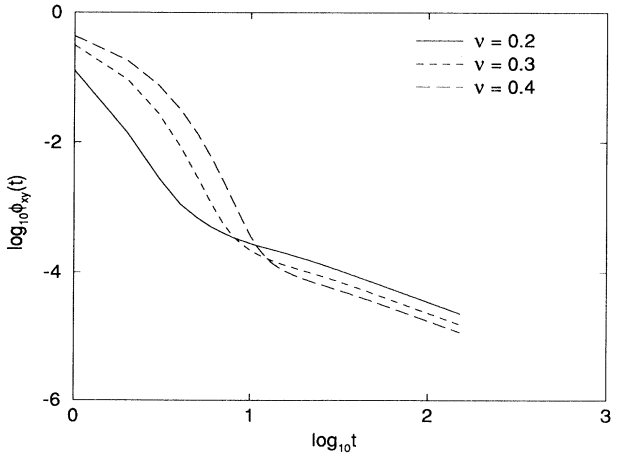

FIG. 1. The normalized stress autocorrelation function ϕ_{xy} of a two-dimensional lattice-gas fluid at a dimensionless kinematic viscosity of $\nu=0.2$, 0.3, 0.4 as a function of time. Time is expressed in units of the discrete time step in the lattice model.

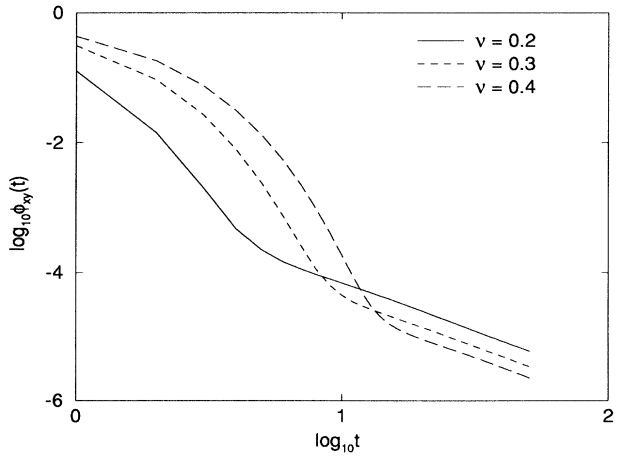

FIG. 3. The normalized stress autocorrelation function ϕ_{xy} of a three-dimensional lattice-gas fluid at a dimensionless kinematic viscosity of $\nu=0.2$, 0.3, 0.4 as a function of time. Time is expressed in units of the discrete time step in the lattice model.

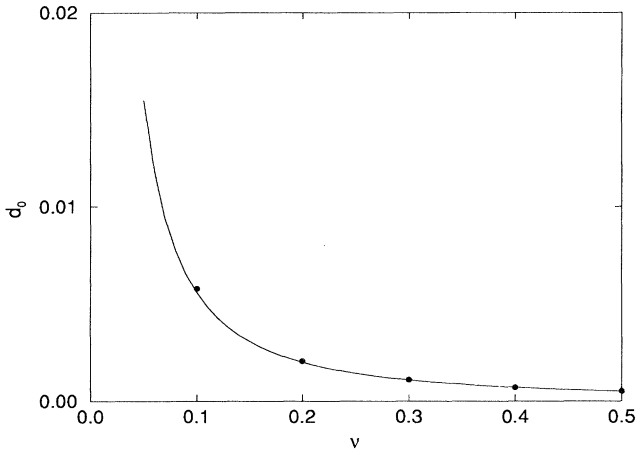

FIG. 4. The tail coefficient of the stress ACF for a three-dimensional lattice-gas fluid, as a function of the dimensionless kinematic viscosity ν. The points are the results of simulations of the lattice-Boltzmann model, while the drawn curve line corresponds to the prediction of the mode-coupling theory.

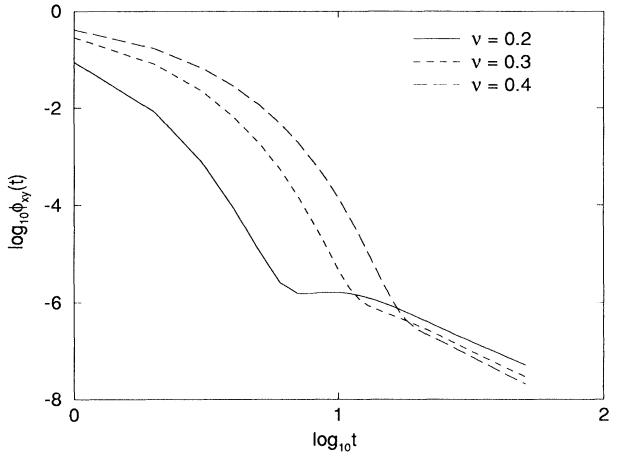

FIG. 5. The normalized stress autocorrelation function ϕ_{xy} of a three-dimensional lattice-gas fluid, with a symmetric stress, at a dimensionless kinematic viscosity of $\nu=0.2$, 0.3, 0.4 as a function of time. Time is expressed in units of the discrete time step used in the lattice model.

obtained from the intercept of $t^{3/2}\phi_{xy}(t)$ in the limit $1/t\rightarrow 0$.

Figure 4 shows a comparison of the tail coefficient obtained from the simulations, with the corresponding model-coupling prediction [Eq. (1)]. As can be seen from this figure, there is again quantitative agreement between mode-coupling theory and the simulation results. The discrepancy is of the order of one percent, somewhat greater than in two dimensions, however, in the three dimensions there is a larger error associated with the extrapolation procedure because the data do not extend to such long times.

In the three-dimensional system we have also computed to what extent the long-time tail in the stress ACF changes the "bare" kinematic viscosity ν which was computed at the Boltzmann level. This is done by using the Green-Kubo formula for the viscosity [15]

$$\nu_{\text{hydro}}(t)\sim\tfrac{1}{2}\phi_{xy}(0)+\sum_{t'=1}^{t}\phi_{xy}(t')\ . \tag{9}$$

Asymptotically, $\nu_{\text{hydro}}(t)\sim t^{1/2}$ [from Eq. (1)], and in this way extrapolation was performed to find $\nu_{\text{hydro}}=\lim_{t\rightarrow\infty}\nu_{\text{hydro}}(t)$. The result of this calculation is shown in Table I. Note that the algebraic long-time tail results in a small renormalization of the viscosity. The same calculation was not performed in two dimensions, because the renormalized kinematic viscosity diverges in that case.

TABLE I. The relative effect of the hydrodynamic long-time tail in the stress autocorrelation function on the dimensionless viscosity in three dimensions.

ν	$(\nu_{\text{hydron}}-\nu)/\nu$
0.1	0.171
0.2	0.079
0.3	0.053
0.4	0.041
0.5	0.033

In the three-dimensional system we have also compared different initial stresses. In the examples discussed above, the initial stress corresponds to a perturbation of the velocity of an arbitrary node **r**. This is the correct way to impose kinematic stress, because such stresses are in practice due to local fluctuations in the velocity distributions of particles: these fluctuations are uncorrelated. In this way the total momentum in the fluid was also increased, see Eq. (2). For the sake of comparison we also considered a symmetric initial stress such that no momentum was introduced in the fluid . The resulting stress ACF for this system is shown in Fig. 5. We see that the exponent of the algebraic long-time tail is now -2.5. This is not surprising because the leading contribution to the tail is canceled exactly. Although these tails clearly do not correspond to the decay of the kinematic part of the stress tensor [16], they may correctly describe the decay of the symmetric stresses, such as those caused by interparticle forces. In the two-dimensional system the computation was also performed, and the exponent of the algebraic long-time tail was found to be -2.

IV. CONCLUSIONS

We have computed the stress ACF of a lattice-Boltzmann fluid and compared the results with the mode-coupling theory. We find that both the exponent of the algebraic long-time tail $(-d/2)$ and its amplitude (d_0) are in essentially quantitative agreement with the mode-coupling theory The computation of the time dependent viscosity in three dimensions shows that, at least for the simple lattice-gas model studied in this work,

the hydrodynamic long-time tail of the stress ACF results in a small correction to the Boltzmann prediction of the kinematic viscosity.

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