Evidence for Faster-than-t^{-1} Decay of the Velocity Autocorrelation Function in a 2D Fluid

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(Received 19 November 1990)

We report very accurate simulations of the velocity autocorrelation function of a tagged particle in a two-dimensional lattice-gas cellular automaton. We observe, for the first time, that the hydrodynamic long-time tail of the velocity autocorrection function decays significantly faster than t^{-1}. The simulation results are compatible with the predictions of self-consistent mode-coupling theory. However, observation of a true $(t\sqrt{\ln t})^{-1}$ decay, which is predicted by the latter theory as $t\to\infty$, would require simulations that are 15 orders of magnitude longer than is currently feasible.

PACS numbers: 61.20.Lc, 05.20.Dd, 05.40.+j, 05.60.+w

Twenty years ago, Alder and Wainwright[1] reported the observation, by molecular-dynamics simulation, of a long-time tail in the velocity autocorrelation function (VACF) of a tagged particle in a classical fluid. Their findings indicated that, at long times, the VACF decays algebraically rather than exponentially. In two-dimensional systems the simulation data were consistent with an asymptotic decay of the VACF $\sim t^{-1}$. Alder and Wainwright gave a simple "hydrodynamic" interpretation of this behavior, and suggested that the algebraic decay could be explained in terms of the slow decay of the hydrodynamic vortex that is induced in the fluid by the initial motion of the tagged particle. If the diffusive motion of this particle is characterized by the diffusion constant D, and the vorticity diffusion of the hydrodynamic flow field by the kinematic viscosity ν, the VACF is expected to a decay as

$$\langle v_x(0)v_x(t)\rangle \sim 1/(D+\nu)t\,, \tag{1}$$

which is in agreement with the Alder-Wainwright simulation results. This theoretical interpretation was subsequently rederived and extended by several authors, using mode-coupling theory[2] and kinetic theory.[3] However, it was already realized at the time that the above theoretical description of the long-time tails is internally inconsistent for two-dimensional fluids. The reason is the following: The diffusion constant D as used in Eq. (1) is defined by the Green-Kubo relation

$$D=\lim_{t\to\infty}\int_0^t \langle v_x(0)v_x(s)\rangle ds\,. \tag{2}$$

As a consequence, D diverges as $\ln t$ if the VACF decays as t^{-1}. In other words, the 2D diffusion constant does not exist. Moreover, the viscosity, which is the integral of the stress-stress correlation function, is also expected to diverge. Alder, Wainwright, and Gass tried to overcome this difficulty by introducing time-dependent transport coefficients,[4] so that the hydrodynamic description could be made self-consistent. In this picture, the decay of the VACF is slightly faster than t^{-1}, namely,

$$\langle v_x(0)v_x(t)\rangle \sim 1/t\sqrt{\ln t}\,. \tag{3}$$

Unfortunately, the corrections to the t^{-1} decay were too weak to be observed in the simulations of Alder, Wainwright, and Gass. More recent simulations of the VACF of a 2D fluid did corroborate the existence of the t^{-1} tail,[5,6] but faster decay could still not be seen.

Recently, lattice-gas cellular automata (LGCA) have been studied as an alternative model for a hydrodynamic fluid.[7,8] In LGCA's, time and space are discrete. Particles are situated at lattice nodes, and can move in a unit time step Δt to neighboring lattice nodes at a distance Δl. Hence particles can only have velocities directed along connecting lattice links. No two particles can be at the same node with the same velocity. Particles that arrive at the same lattice site may collide, under the condition that the total momentum and the number of particles is conserved. It has been demonstrated[7,8] that it is possible to choose the LGCA model such that the equation that governs the time evolution of the distribution function of such a lattice gas becomes equivalent to the Navier-Stokes equation for an incompressible fluid. The specific model used in this Letter is the two-dimensional Frisch-Hasslacher-Pomeau- (FHP-) III model, as defined in Ref. 9. In this model, particles move on a triangular lattice and can therefore have six different velocities. Moreover, a particle can have zero velocity. For the FHP-III model, mode-coupling theory[10,11] predicts that the long-time tail of the VACF of a tagged particle is of the following form:

$$\langle v_x(0)v_x(t)\rangle = \frac{3\sqrt{3}}{784\pi}\left[\frac{1-d}{d}\right]\frac{1}{(D+\nu)t}\,, \tag{4}$$

where d is the density defined as the average number of particle per link ($0\le d\le 1$), and where we have introduced Δl and Δt as units of length and time, respectively. The VACF is defined as the time average of the product $v_x(0)v_x(t)$ along any possible trajectory of the tagged particle. Clearly, in a simulation the statistical accuracy can be improved by averaging over different trajectories. In LGCA's, this can be done very efficiently because of the indistinguishability of lattice-gas particles. This makes it possible to simultaneously compute *all* tagged-

particle trajectories rather than just one. In this way a much improved estimate of the VACF is obtained.[12,13] Compared to "single-trajectory" methods, the gain in computing speed amounts to more than 6 orders of magnitude. Recently, we have used the above scheme to compute the VACF of a three-dimensional lattice gas[10] and found the results of these simulations to be in good quantitative agreement with the predictions of the corresponding mode-coupling theory.[11] In this 3D mode-coupling theory, the "Boltzmann" estimate for the transport coefficients D and v are used. Both in 2D and in 3D the differences between the Boltzmann estimates for D and v and the exact Green-Kubo expressions are almost exclusively due to the hydrodynamic long-time tails. In 3D this difference is very small ($\approx 0.1\%$). In contrast, as explained above, we should expect that in 2D the corrections to the Boltzmann transport coefficients will not just change the long-time tail quantitatively, but even qualitatively. However, in spite of the improved accuracy, the long-time corrections to the t^{-1} tail in 2D lattice gases could not be observed.[13] There are two possible explanations why corrections to the t^{-1} decay have not been observed thus far: (1) The simulations were not yet sufficiently accurate, or (2) the physical reasoning behind self-consistent mode coupling is seriously flawed. The latter conclusion would be worrisome because self-consistent mode-coupling theories have been applied to many problems in physics. It is therefore of considerable interest to test the prediction of self-consistent mode-coupling theory in the present "prototypical" example. In the present Letter we present the results of extensive simulations that were aimed specifically at estimating the deviation (if any) from t^{-1} decay in the VACF.

Runs were done for a range of densities and correlation time intervals. In all cases we did choose our systems sufficiently large so that no particle could cross the periodic box within the correlation time interval. A correlation of $\langle v_x^2\rangle(1-d)/N$ was added to the VACF (N is the total number of particles) to correct for finite-size effects associated with the fact that we constrain the total momentum of the system to vanish. In order to increase the statistical accuracy (at the expense of time resolution), we averaged the quantity of interest $[t\langle v_x(0)v_x(t)\rangle]$ over a time window of n steps centered around t, where n was varied from 100 to 400. All the simulations reported here were carried out at densities ranging from $d=0.70$ to $d=0.80$. The reason is that for *lower* densities the statistics of the simulations becomes worse, while for *higher* densities the deviations from t^{-1} decay are expected to be less pronounced.[14] The simulations reported here took some 100 hours on a NEC-SX2 supercomputer.

Figure 1 shows a log-log plot of the velocity autocorrelation function of a tagged particle in a lattice gas at density $d=0.80$ (solid curve). The dashed curve indicates the estimated statistical error. Time is in units of

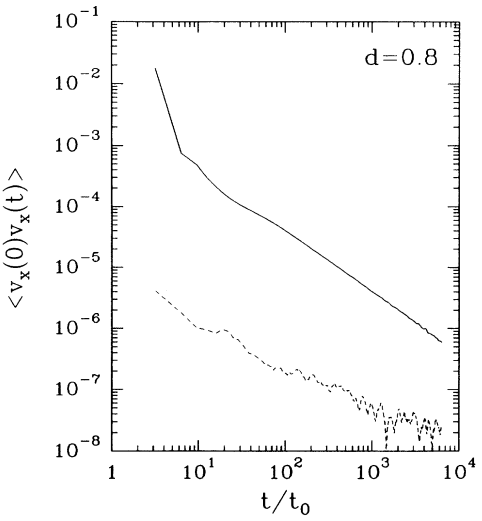

FIG. 1. Typical example of the simulation results for the velocity autocorrelation function of a tagged particle in a LGCA (solid curve). The density is equal to $d=0.8$. t_0 (see text) is equal to 0.31 in units of Δt. Note that the error (dashed curve) decreases with increasing time to a value of order 10^{-8}. The error was estimated as the standard deviation in the results of nine independent simulations. However, the error bars on our data points (see Figs. 2 and 3) should not be considered independent, as the statistical fluctuations at different times are strongly correlated.

the mean free time t_0, which is determined by the initial exponential decay $\sim\exp(-t/t_0)$. Figure 1 shows that we can measure the VACF over more than three decades in time with an almost constant signal-to-noise ratio. Note that the VACF can be measured up to 6000 mean free times, without interference of the periodic images. In order to observe any faster-than-t^{-1} decay it is more convenient to plot the VACF multiplied by t on a linear scale, as shown in Fig. 2. In this figure, we plotted the velocity autocorrelation function multiplied by the time t, for densities $d=0.70$ (curve A), $d=0.75$ (curve B), and $d=0.80$ (curve C). If the VACF deviates from the predictions of lowest-order mode-coupling theory, i.e., that version of the mode-coupling prediction (4) in which the transport coefficients D and v take the (time-independent) Boltzmann values, the simulation results should differ significantly from the dashed horizontal lines in Fig. 2. This is indeed observed for all densities studied. Moreover, if the VACF decays faster than t^{-1}, the measured curves should have a negative slope. This behavior is clearly observed in Fig. 2 at density $d=0.75$ and 0.80. To our knowledge this is the first time that a decay faster than t^{-1} of the VACF is demonstrated in a simulation. Still, the effect is very small, both on an absolute and on a relative scale. This is clearly demonstrated by the results at a density $d=0.70$ (Fig. 2, curve A), where the faster-than-t^{-1} decay is almost unobservable due to the relatively large statistical errors.

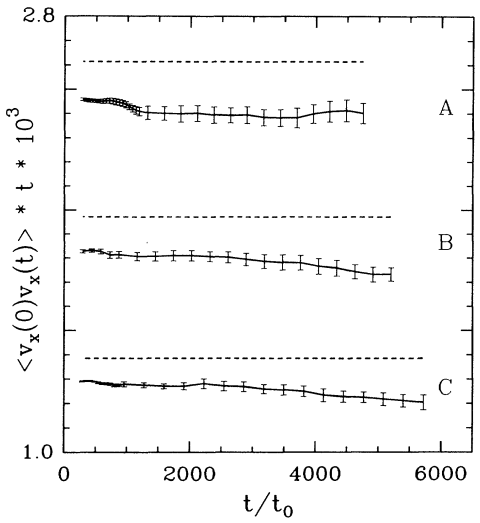

FIG. 2. Velocity autocorrelation function of a tagged particle multiplied by time, for densities $d=0.70$ (curve A), $d=0.75$ (curve B), and $d=0.80$ (curve C). The solid curves denote the simulation data (with error bars). The fact that all these curves have, on average, a negative slope is indicative of faster-than-t^{-1} decay. The dashed lines are the predictions of lowest mode-coupling theory [Eq. (4)], using the (known) Boltzmann transport coefficients. t_0 is equal to 0.38, 0.35, and 0.31 for densities $d=0.70$, 0.75, and 0.80, respectively, in units of Δt.

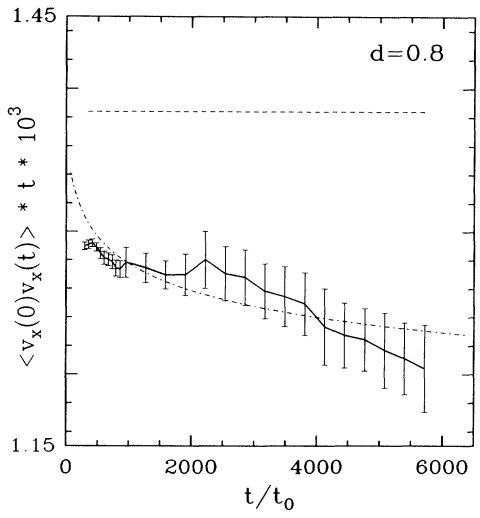

FIG. 3. Velocity autocorrelation function of a tagged particle multiplied by time, for density $d=0.8$. Solid curve: simulation data (same as Fig. 2); dashed line: prediction of lowest-order mode-coupling theory; dash-dotted curve: prediction of mode-coupling theory with the true time-dependent transport coefficients. The true diffusion coefficient is obtained from the measured VACF; the true viscosity is estimated from lowest-order mode-coupling theory.

Let us next consider a comparison with higher-order mode-coupling theory. In that case, we should use the mode-coupling prediction (4) with the "true" time-dependent transport coefficients, rather than the Boltzmann estimate. The time-dependent diffusion coefficient can be obtained directly from the measured VACF by integration. In the same manner, the true viscosity is proportional to the integral of the stress-stress correlation function. However, the latter function could not be measured in the simulation with sufficient accuracy. We have therefore used the prediction of lowest-order mode-coupling theory for the long-time behavior of the stress-stress correlation function.[14] This prediction was also derived by Kadanoff, McNamara, and Zanetti,[15] and was in agreement with their simulations. Hence, lowest-order mode-coupling theory should give a sufficiently accurate estimate of the time-dependent viscosity to compare with the present simulations. Having thus obtained the time-dependent D and ν, we can compare the measured VACF with the higher-order mode-coupling theory. This comparison is shown in Fig. 3 for one particular density ($d=0.80$). The solid curve with error bars is the simulation result; the dashed line is the prediction of lowest-order mode-coupling theory (both are the same as in Fig. 2). The dash-dotted curve is the prediction of higher-order mode-coupling theory. From this figure we can draw two important conclusions. First, the magnitude of the tail predicted by higher-order mode-coupling theory is in agreement with the simulations. Second, the faster-than-t^{-1} decay observed in the simulation is compatible with the faster decay due to the use of the true transport coefficients in (4). We stress that the latter is the basic idea behind self-consistent mode-coupling theory. At the other two densities that we studied ($d=0.70$ and 0.75) the simulation results also agreed within the estimated statistical error with the predictions of higher-order mode-coupling theory. It would be interesting to determine whether the functional form of the observed VACF is compatible with the *full* self-consistent prediction (3). However, in order to observe the latter behavior, the divergent contributions to D and ν should exceed the Boltzmann values. We estimate that this happens after some 10^{20} mean free times. This suggests that it will be effectively impossible to establish true $(t\sqrt{\ln t})^{-1}$ decay in the present class of LGCA's. This asymptotic behavior should be more pronounced in model systems where the relative amplitude of the hydrodynamic long-time tail is much larger, for example, in a 2D system of colloidal particles.[16] However, in that case we cannot employ the algorithm that makes efficient calculation of the VACF in LGCA's feasible.

We wish to acknowledge stimulating and useful discussions with Berni Alder, Matthieu Ernst, and Toyoaki Naitoh. The work of the FOM Institute is part of the scientific program of FOM and is supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO). Computer time on the NEC-SX2 at the

Nationaal Lucht- en Ruimtevaartkundig Laboratorium (NLR) was made available through a grant by the Nationaal Fonds Supercomputers (NFS).

[1]B. J. Alder and T. E. Wainwright, Phys. Rev. A **1**, 18 (1970).

[2]M. H. Ernst, E. H. Hauge, and J. M. J. van Leeuwen, Phys. Rev. A **4**, 2055 (1971).

[3]J. R. Dorfman and E. G. D. Cohen, Phys. Rev. Lett. **25**, 1257 (1970).

[4]B. J. Alder, T. E. Wainwright, and D. Gass, Phys. Rev. A **4**, 233 (1971).

[5]J. J. Erpenbeck and W. W. Wood, Phys. Rev. A **26**, 1648 (1982).

[6]M. E. Colvin, A. J. C. Ladd, and B. J. Alder, Phys. Rev. Lett. **61**, 381 (1988).

[7]U. Frisch, B. Hasslacher, and Y. Pomeau, Phys. Rev. Lett. **56**, 1505 (1986).

[8]U. Frisch, D. d'Humières, B. Hasslacher, P. Lallemand, Y. Pomeau, and J.-P. Rivet, Complex Systems **1**, 649 (1987).

[9]D. d'Humières and P. Lallemand, Complex Systems **1**, 599 (1987).

[10]M. A. van der Hoef and D. Frenkel, Phys. Rev. A **41**, 4277 (1990).

[11]M. H. Ernst, Physica (Amsterdam) **47D**, 198 (1991).

[12]D. Frenkel, in *Proceedings of the Workshop on Cellular Automata and Modelling of Complex Physical Systems, Les Houches, France, 21–28 February 1989,* edited by P. Manneville (Springer, Berlin, 1989).

[13]D. Frenkel and M. H. Ernst, Phys. Rev. Lett. **63**, 2165 (1989).

[14]T. Naitoh, M. H. Ernst, and J. W. Dufty, Phys. Rev. A **42**, 7187 (1990).

[15]L. P. Kadanoff, G. R. McNamara, and G. Zanetti, Phys. Rev. A **40**, 4527 (1989).

[16]M. A. van der Hoef, D. Frenkel, and A. J. C. Ladd (to be published).