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COMPUTER SIMULATIONS OF LONG-TIME TAILS: WHAT'S NEW?

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ABSTRACT

Twenty five years ago Alder and Wainwright discovered, by simulation, the 'long-time tails' in the velocity autocorrelation function of a single particle in fluid [1]. Since then, few qualitatively new results on long-time tails have been obtained by computer simulations. However, within the framework of a lattice-gas simulation, we recently developed a technique that makes it possible to 'measure' such velocity autocorrelation functions with an efficiency that is at least a factor 10^6 higher than can be achieved with conventional techniques. This method opens the way for making comparisons with the predictions of mode-coupling theory of long-time tails, to an accuracy which was hitherto not possible. In this paper we describe this method, and review the results on long-time tails that have been obtained. In particular, we present evidence that the functional form of the long-time tail in a two-dimensional fluid is qualitatively different from the simple power law observed by Alder-Wainwright. Such 'faster-than-t^{-1}' decay is in agreement with the predictions of 'self-consistent' mode-coupling theory.

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I. INTRODUCTION

The long-time tail in the velocity autocorrelation function (VACF) of a single particle in a fluid, is an almost 'proto-typical' example of a phenomenon which can be described by mode-coupling theory. The origin of the long-time tail lies in the fact that the initial motion of the tagged particle will set up a hydrodynamic flow field in the surrounding fluid ('shear modes'), which recirculates around the particle and exerts a slowly decaying force on it. From this follows, if diffusive motion of the particle is neglected, that the VACF of the particle decays as $(\nu t)^{-d/2}$, where d is the dimension of the system. Mode-coupling theory accounts for the effect of the diffusion of the single particle, i.e. the coupling of the shear and diffusive modes; the fact that these modes couple means that we can include the effect of diffusion simply by replacing ν by $D + \nu$, where D is the diffusion coefficient. Hence the VACF of a diffusive particle decays for long-times as $((D + \nu)t)^{-d/2}$. Long-time tails were discovered 25 years ago in a computer simulation by Alder and Wainwright [1]. This came as a complete surprise, as up to then the decay of the VACF was believed to be exponential, which follows from the 'Molecular Chaos' assumption, which states that the collisions experienced by a molecule in a fluid are uncorrelated. The discovery of the long-time tails had important consequences for the kinetic theory of fluids. Naturally, it demonstrated the breakdown of the 'Molecular Chaos' assumption, which has been one of the key assumptions of kinetic theory for close to a hundred years. Secondly, the interpretation of the tails demonstrated that hydrodynamic arguments can be applied to describe the behavior of microscopic particles, implying that the laws of hydrodynamics seem to be valid down to surprisingly small scales. Thirdly, the long-time tails themselves have important consequences for the transport coefficients, in particular in two dimensions. Namely, in two dimensions, the VACF decays as t^{-1}, which implies that the diffusion coefficient, which can be identified with the integral over the VACF, diverges! Hence, the transport coefficients, as they are normally defined do not exist in 2D. Incidentally, this divergence has also its effect on the validity of the theoretical prediction of the long-time tail itself, as in mode-coupling theory it is assumed that the diffusion coefficient is constant in time. The theory can be modified for this, by allowing the two-dimensional diffusion coefficient to be time dependent, and solve the equations self-consistently. It turns out that this 'self-consistent'

long-time tail decays slightly faster than t^{-1}: namely as $(\ln t)^{-\frac{1}{2}}t^{-1}$. The existence of these corrections was already recognized by Alder and Wainwright at the time of their discoveries. However, their data were not accurate enough to observe any deviations from t^{-1} decay. In fact, even with the present day supercomputer, the VACF can *still* not be calculated in a molecular dynamics simulation with sufficient accuracy in order to see these corrections. However, we recently developed a method, where the VACF of a particle in a lattice gas can be calculated at least a factor 10^6 more efficient. Although lattice gases are perhaps not so 'realistic' as molecular dynamics models, they do contain all the essential physics underlying the long-time tails, namely hydrodynamic behavior of the collective system, and diffusive motion of the individual particles. In this contribution we would like to present which 'new' results on long-time tails have been obtained using this method. The organization of the paper is as follows. In section 2 we give a very simple outline of the derivation of long-time tails from mode-coupling theory, and especially show how one obtains the self-consistent long-time tail. In section 3 we show the essentials of the efficient method by which the VACF can be calculated in a lattice-gas simulation. What we hope to show is that the formalism is extremely simple, and moreover we expect that it can be applied to many other systems. In section 4 we show results obtained on long-time tails in two, three and four dimension. And finally in section 5 we show evidence for a faster-than-t^{-1} decay of the VACF in two dimensions.

II. MODE-COUPLING THEORY

II.1 Self-Diffusion

As a preliminary, we briefly define the various quantities of interest in the the study of self-diffusion, mainly to introduce the time-dependent transport coefficients in their proper context. The position and velocity of a tagged particle in a fluid are given by $\mathbf{r}(t)$ and $\mathbf{v}(t)$ respectively. We define the mean-square displacement $\Delta(t)$ of the tagged particle by

$$\Delta(t) = \frac{1}{2d}\langle(\mathbf{r}(t) - \mathbf{r}(0))^2\rangle \,,$$

and the velocity autocorrelation function $\phi(t)$ by

$$\phi(t) = \langle v_x(0)v_x(t)\rangle \ .$$

In the above equation (and in what follows),$\langle\cdots\rangle$ means a statistical average, and d is the dimension of the system. By making use of the identity $\mathbf{r}(t) - \mathbf{r}(0) = \int_0^t \mathbf{v}(s)ds$, we get the well-known relation between $\Delta(t)$ and $\phi(t)$:

$$\Delta(t) = \int_0^t ds' \int_0^{s'} ds\phi(s) \ . \tag{1}$$

If the motion of the particle is purely diffusive, the displacement statistics of the particle is Gaussian, i.e. if the tagged particle starts at time $t = 0$ at position $\mathbf{r}_0$, its distribution function $P(\mathbf{r}, t)$ is given by

$$P(\mathbf{r}, t) = (4\pi Dt)^{-d/2} \exp\left[-\frac{(\mathbf{r} - \mathbf{r}_0)^2}{4Dt}\right] \ , \tag{2}$$

which defines the self-diffusion coefficient D. The mean-square displacement will then be equal to

$$\Delta(t) = \frac{1}{2d} \int P(\mathbf{r}, t)(\mathbf{r} - \mathbf{r}_0)^2 d\mathbf{r} = Dt \ . \tag{3}$$

Note that normally diffusive behavior is only achieved after longer times. Then the diffusion coefficient can be expressed as

$$D = \lim_{t\to\infty} \frac{\Delta(t)}{t} = \lim_{t\to\infty} \int_0^t ds\phi(s) \ .$$

In last identity use has been made of the assumption that $\phi(t)$ is invariant under translation in time, and decays faster than t^{-1}. As has been discussed in the introduction, the simple picture of diffusion given above need not always be true. In particular, the mean-square displacement might be growing faster than linearly in time, and hence the distribution function (2) is not correct. However, if these deviations from purely diffusive behavior are on a much larger time scale than the diffusion phenomenon itself, we might use the same form for the probability distribution function, but with D replaced by a time-dependent $D(t)$, which varies on long time scales only. It follows then that the mean-square displacement is equal to

$$\Delta(t) = D(t)\, t \tag{4}$$

Eqn. 4 will be used later in the explanation of the self-consistent long-time tails.

II.2 Mode-Coupling Approach to Long-Time Tails

In mode-coupling theory, the basic idea is that the modes which describe the decay of particle's initial velocity (shear modes), and the modes which describe the particle displacement (diffusive modes), couple. This theory is completely based on the assumption that hydrodynamic arguments alone are enough to explain the long-time tails. Below we only briefly sketch the basic idea; for a complete and more rigorous derivation we refer to the original paper by Ernst *et al* [2]. Suppose the particle starts at time $t = 0$ at coordinate $\mathbf{r}_0$, with velocity $\mathbf{v}_0$. We do not yet specify the type of particle. Now the crucial assumption is that, after long enough time t, the velocity of the particle will be equal to the local fluid velocity, so that we can write for the average velocity of the particle at time t:

$$\mathbf{v}(t) = \int_V P(\mathbf{r},t)\mathbf{u}(\mathbf{r},t)d\mathbf{r} \,, \tag{5}$$

where $P(\mathbf{r},t)$ is the single-particle probability density and $\mathbf{u}(\mathbf{r},t)$ is the hydrodynamic flow field. We assume that the evolution of $\mathbf{u}$ is described by the Navier-Stokes equation of an incompressible fluid, in the low-Reynolds number limit:

$$\frac{\partial \mathbf{u}(\mathbf{r},t)}{\partial t} = -\frac{1}{\rho}\nabla P + \nu\nabla^2\mathbf{u}(\mathbf{r},t) \,, \tag{6}$$

$$\nabla \cdot \mathbf{u} = 0 \,. \tag{7}$$

We are interested in the evolution of the flow field, caused by the initial motion of the particle, which means that one needs to specify the initial flow field at the particle boundary, which requires detailed knowledge of the nature of the particle (size, shape, stick or slip boundary conditions). Therefore we make the assumption that, after long enough times, the flow field that gives rise to the long-time tails is determined by the *amount* of momentum that is introduced initially into the fluid, and not by *how* precisely it was introduced into the fluid. In that case, we may assume that the flow field that develops from the initial motion of a particle with velocity $\mathbf{v}_0$ and mass M, will be equivalent to the flow field that develops from a small volume element of fluid δV around $\mathbf{r} = \mathbf{r}_0$, with an initial flow field $\mathbf{u}_0$, such that $M\mathbf{v}_0 = \rho\mathbf{u}_0\delta V$, and where the fluid is at rest outside the volume element. If we can make δV arbitrary small, the initial conditions becomes

$$\mathbf{u}(\mathbf{r},0) = \frac{M\mathbf{v}_0}{\rho}\delta(\mathbf{r}-\mathbf{r}_0) .$$

If the pressure term in (6) could be neglected, then what remains is a simple diffusion-type equation, with an initial delta condition. The solution would be

$$\mathbf{u}(\mathbf{r},t) = \frac{M\mathbf{v}_0}{\rho}(4\pi\nu t)^{-d/2}\exp\left[-\frac{(\mathbf{r}-\mathbf{r}_0)^2}{4\nu t}\right] . \tag{8}$$

However, we cannot neglect the pressure term. If included, a standard calculation (in Fourier space) shows that the relevant part of the velocity field will be equal to (8), only $(d-1)/d$ smaller. Next we assume that the particle has purely diffusive motion, so $P(\mathbf{r},t)$ is given by (2). Hence, the integral (5) is now simply over the product of two Gaussians (which is the real-space analogy of 'mode-coupling')

$$\mathbf{v}(t) = \int_V P(\mathbf{r},t)\mathbf{u}(\mathbf{r},t) =$$

$$\left(\frac{d-1}{d}\right)\frac{M\mathbf{v}_0}{\rho}((4\pi t)^2 D\nu)^{-d/2}\int_V d\mathbf{r}\exp\left[-\frac{(\mathbf{r}-\mathbf{r}_0)^2}{4Dt}-\frac{(\mathbf{r}-\mathbf{r}_0)^2}{4\nu t}\right]$$

$$= \left(\frac{d-1}{d}\right)\frac{M\mathbf{v}_0}{\rho}(4\pi(D+\nu)t)^{-d/2} .$$

Multiplying by $\mathbf{v}_0$ and taking the average over all initial velocities gives

$$\phi(t) = \left(\frac{d-1}{d}\right)\frac{M\phi(0)}{\rho}[4\pi(D+\nu)t]^{-d/2} . \tag{9}$$

We note that this is a very simple way to demonstrate the basic idea. In a more sophisticated approach [2] one uses the linearized Navier-Stokes equations, in which the fluid is not strictly incompressible, but where the density has small fluctuations. In that case, the initial momentum decays via $d-1$ shear modes, which gives a velocity field equivalent to the one used in the analysis above, and via one sound mode; the latter does *not* couple to the diffusive modes, hence one gets te same result (9) for the long-time tail. Note that the expression derived above should be regarded as the lowest-order version of mode-coupling theory, as it assumes that the transport coefficient take their time-independent 'Boltzmann' values. However, as discussed before, the long-time tails will modify these transport coefficient, which could be taken into account by higher-order versions of the theory. Next, we discuss an higher-order version in which the use of non-Boltzmann estimates of the transport coefficient will not only modify the amplitude of the long-time tail, but also its functional form.

II.3 Self-Consistent Mode-Coupling Theory

From mode-coupling theory it follows that, in two dimensions, the long-time tail is given by

$$\phi(t) = \frac{b}{(D+\nu)\,t}\,, \tag{10}$$

where

$$b = \frac{M\phi(0)}{8\pi\rho}\,.$$

Eqn. 10 is valid if the transport coefficients D and ν are asymptotically reaching a constant value. Let us assume, for the moment, that ν is indeed constant. However, if we make the same asumption for D, we end up with an inconsistency: D is related to the mean-square displacement by (3); the mean-square displacement in turn is equal to the double integral over the VACF (1). If the VACF decays as t^{-1}, then the double integral grows as $t \ln t$ and hence the diffusion coefficient diverges as $\ln t$ for $t \to \infty$; consequently the long-time tail (10) has a functional form different from t^{-1}. As was first suggested by Wainwright *et al* [3], a self-consistent picture can be obtained by introducing a time-dependent diffusion coefficient; the mode-coupling expression for the long-time tail then becomes

$$\phi(t) = \frac{b}{(D(t)+\nu)t}\,.$$

If we combine this with Eqs. (1) and (4), we get the differential equation

$$\frac{d^2(D(t)t)}{dt^2} = \frac{b}{D(t)t + \nu t}\,. \tag{11}$$

The asymptotic solution of this equation, at sufficiently long-times that $D(t) \gg \nu$, is

$$D(t) = \sqrt{2b}\,\sqrt{\ln t}\,, \tag{12}$$

whereas a naive (inconsistent) picture would give $D(t) \sim \ln t$. The self-consistent form of the diffusion coefficient (12) corresponds to a velocity autocorrelation function which does not decay as t^{-1}, but slightly faster:

$$\phi(t) = \sqrt{\frac{b}{2}}\left(\frac{1}{t\sqrt{\ln t}}\right)\,. \tag{13}$$

In the above we made the assumption of a time-independent viscosity. How-

ever, the viscosity is equal to the time-integral over the stress-stress autocorrelation function, which has a similar long-time tail. Hence, the viscosity should be made time-dependent as well, and one can derive an equation analogous to (11), and solve the two coupled differential equations. However the asymptotic form (13) remains the same. Finally we note that the same asymptotic form for the decay of the VACF has been derived *ab initio*, by summing all the mode-coupling contributions to the long-time tail [4].

III. COMPUTATIONAL TECHNIQUES

III.1 Introduction

The velocity autocorrelation function of a tagged particle is can be defined as the time average of the product $v_x(0)v_x(t)$ along any possible trajectory of the tagged particle, viz. (for time-discrete systems)

$$\langle v_x(0)v_x(t)\rangle = \lim_{T\to\infty} \frac{1}{T} \sum_{n=0}^{T} v_x(n)v_x(t+n) \; . \tag{14}$$

In a conventional molecular dynamics simulation, a single particle has a well defined trajectory, and (14) can be applied directly. In a simulation run of length T, the statistical error in the the measured value of $\langle v_x(0)v_x(t)\rangle$ will be proportional to $T^{-1/2}$ and is independent of t, for t larger than the initial decay time [5]. As fluctuations decay, the VACF decreases for 'long' times. As a consequence, the signal-to-noise ratio is decreasing as well, and lengthy simulations are required to study the long-time behavior of the VACF by use of molecular dynamics. In a lattice-gas simulation the situation is different. In such a simulation particles move on a lattice and interact in point collisions at the nodes. The usual collision rules in a lattice gas do not specify which particle ends up in which final state. Hence, in a sense, the particles lose their identity during a collision. This could be remedied by having special collision rules for tagged and untagged particles, for instance in which the tag is attached to one of the outgoing particles by random selection. In this way, (14) can be applied again, but also in this case brute force is required to observe the VACF for longer times [6]. However, the lattice structure makes a more efficient approach possible [7, 8]; the basic idea is that on a lattice one can

consider *all* possible tag distributions at once, instead of only one at a time, as in (14). Before we describe this method we should first introduce the lattice-gas model in more detail.

III.2 Lattice Gas Cellular Automata

Lattice gas cellular automata (LGCA), introduced by Frisch, Hasslacher and Pomeau [9] in 1986, can best be viewed as a discrete molecular dynamics. The idea behind the model is that the hydrodynamic behavior of a system does not crucially depend on the details of the collisions between the individual particles. It is therefore well possible to use a model with less 'realistic', but computationally more efficient collisions, which might still give correct hydrodynamic behavior. In lattice gas cellular automata this idea is carried out *in extremum*, making it the simplest known model of a fluid. In such models, time and space are discretized, thus the particles are confined to a lattice, the state of which is only defined at discrete times with intervals Δt. As a consequence, the particles also have discrete velocities such that the particles move to a neighboring lattice site in one time step. The b possible velocities are given by the set $\{\mathbf{c}_1, \mathbf{c}_2, ..., \mathbf{c}_b\}$, each of which should satisfy the constraint

$$\mathbf{r}' = \mathbf{r} + \mathbf{c}_i \Delta t \ ,$$

where $\mathbf{r}$ and $\mathbf{r}'$ must be neighboring lattice sites. An exclusion rule is imposed which states that no two particles with the same velocity can occupy the same lattice site at the same time. One can imagine a particle with velocity $\mathbf{c}_i$ as to 'occupying' the i-th link of the site connecting a neighboring site. The state of the automaton at a certain time t can be completely determined by the integer value of $n_i(\mathbf{r}, t)$, which is equal to 1(0) if a particle is present(absent) on site $\mathbf{r}$ with velocity $\mathbf{c}_i$. The local microscopic mass density $\tilde{\rho}$ is then defined by

$$\tilde{\rho}(\mathbf{r}, t) = \sum_{i=1}^{b} n_i(\mathbf{r}, t) \ , \tag{15}$$

and the local microscopic velocity field $\tilde{\mathbf{u}}(\mathbf{r}, t)$ is defined by

$$\tilde{\rho}(\mathbf{r}, t)\, \tilde{\mathbf{u}}(\mathbf{r}, t) = \sum_{i=1}^{b} n_i(\mathbf{r}, t)\, \mathbf{c}_i \ . \tag{16}$$

The update of the lattice at every time step then consists of allowing for a

simultaneous propagation of all particles to neighboring lattice sites, followed by a collision at each site, which consists of a redistribution of the particles among the possible velocities, such that the number of particles ($=\tilde{\rho}$) and the momentum ($=\tilde{\rho}\tilde{\mathbf{u}}$) at that site is conserved. The macroscopic behavior of these systems can be derived by applying the conventional procedures of statistical mechanics, i.e. ensemble averaging followed by a Chapman-Enskog type expansion; the resulting macroscopic equations of motion are, in a well-defined limit, equal to the Navier-Stokes equations, provided that the lattice has sufficiently high symmetry and the fluid-flow velocity is much smaller than the speed of sound. For a derivation, see [10]. Hence, lattice gas cellular automata are, like molecular dynamics, a truly *microscopic* model of an atomic fluid, including spontaneous fluctuations and correlations, but has the significant advantage that it is computationally much more efficient than molecular dynamics. Initially it was hoped that, with the use of dedicated hardware, these models could be more powerful than existing techniques to study fluid-flow phenomena, like finite-difference and finite-element schemes. This promise has, thus far, not been fulfilled. However, because of its simplicity, the model is ideally suited to serve as a testing ground for concepts in kinetic theory. Most of the theories of simple fluids, including the theories on long-time tails, can be rederived and tested within the lattice-gas framework [11, 12], with the advantage that the simplicity facilitates both the simulation, and the theoretical evaluation. Verification of a theory in the LGCA fluid model then provides very strong support for the validity of the theory in more realistic fluid models. For calculating the VACF the model is even more suited, as because of the lattice structure one can employ a trick which we describe below.

III.3 Moment-Propagation Method

First we make the observation that in the lattice-gas model described in the previous section, a tagged particle is ill-defined: The collision rules only specify the occupation of the different velocity states after a collision, not the identity of the particles in those states. Hence, with such collision rules any initial labeling of particles is lost after one collision. Of course, it is possible to construct more elaborate rules that keep track of the identity of the colliding particles; for a 'tag-blind' observer, these collisions should not be any different from the ordinary collision rules. An obvious choice is a stochastic

collision rule, in which the tag is distributed among the outgoing particles by random selection. Before we proceed let us be more specific about the time. As discussed in the previous section, at time t there are in fact two distinct moments: just before and just after the collision. In the following, it will be more convenient to denote the time *before* the collision by t^-, and the time *after* the collision by t. We consider the correlation function only at times t, just after the collision. Naturally time is in units of a single lattice-gas time step Δt.

In general, the statistical accuracy of the VACF can be improved by averaging over *different* trajectories, instead of just one, as is done in Eq. (14). The idea of the moment propagation method [7, 8] is that one can consider *all* possible trajectories of the tagged particle at once, with only a moderate increase in computing time. This can be done as follows. The stochastic rules define the probability P_k of each trajectory k of the tagged particle. We define a 'trajectory-averaged' VACF by

$$\langle v_x(0)v_x(t)\rangle_{tr} = \sum_k P_k v_x(0)v_x^k(t)\,, \tag{17}$$

where $v_x^k(t)$ is the velocity of the tagged particle at time t, if it would have followed trajectory k. $\langle\cdot\cdot\rangle_{tr}$ stands for the average over all trajectories. Keeping track of all the trajectories in (17) is very time and memory consuming, but will prove to be not necessary. This is so because in lattice gases, the tagged particle will *not* necessarily have a different final velocity for each different trajectory it could have followed; all the trajectories that end at time t at the same site $\mathbf{r}$ and link i, will have the same final velocity $v_x^k(t) = c_{ix}$ for the tagged particle. Therefore, in Eq. (17) we can combine trajectories that end at the same coordinates $(\mathbf{r}, i)$ at time t:

$$\sum_k P_k\; v_x(0)v_x^k(t) = \sum_{\mathbf{r},i} v_x(0)c_{ix} \sum_{k(\mathbf{r},i)} P_k\,, \tag{18}$$

where the sum over $k(\mathbf{r}, i)$ is over all trajectories k ending at $(\mathbf{r}, i)$ at time t. Clearly this sum

$$\sum_{k(\mathbf{r},i)} P_k = P(\mathbf{r}, i; t) \tag{19}$$

represents the total probability (= via any possible trajectory) of the tagged particle to be at $(\mathbf{r}, i)$ at time t. If the stochastic rules are such that in a 'tagged-particle collision' one of the post-collision particles is tagged at ran-

dom, $P(\mathbf{r}, i; t)$ can be written as the product of the probability $P(\mathbf{r}; t^-)$ of the tagged particle to arrive at site $\mathbf{r}$ before the collision, and the probability that the tagged particle will be scattered into the i-th direction after the collision, viz.

$$P(\mathbf{r}, i; t) = P(\mathbf{r}; t^-) \frac{n_i(\mathbf{r}, t)}{\tilde{\rho}(\mathbf{r}, t)} , \tag{20}$$

where $\tilde{\rho}(\mathbf{r}, t)$ is the total number of particles at site $\mathbf{r}$, defined by (15). Note that $\tilde{\rho}$ is not changed in a collision. This is also true for the probability $P(\mathbf{r}; t^-)$, so we may substitute t^- by t. If we combine equations (17),(18),(19) and (20), and perform the summation over i, we get the following very simple expression

$$\langle v_x(0) v_x(t) \rangle_{tr} = v_x(0) \sum_{\mathbf{r}} P(\mathbf{r}; t) \tilde{u}_x(\mathbf{r}, t) , \tag{21}$$

where $\tilde{u}_x(\mathbf{r}, t)$ is the x-component of the local fluid velocity at site $\mathbf{r}$, defined by (16). Note the resemblance with Eqn. 5 which was used as a starting point in the mode-coupling analysis. The probability $P(\mathbf{r}; t)$ can be constructed from the probabilities at the neighboring sites one time step earlier

$$P(\mathbf{r}, t) = \sum_{i=1}^{b} \left[P(\mathbf{r} - \mathbf{c}_i, t-1) \frac{n_i(\mathbf{r} - \mathbf{c}_i, t-1)}{\tilde{\rho}(\mathbf{r} - \mathbf{c}_i, t-1)} \right] , \tag{22}$$

If the particle starts at $(\mathbf{r}', i')$, at time $t = 0$, the value $P(\mathbf{r}, 1)$ is given by

$$P(\mathbf{r}, 1) = \delta(\mathbf{r} - \mathbf{c}_{i'}, \mathbf{r}') . \tag{23}$$

This also implies that $v_x(0)$ is equal to $c_{i'x}$. Equations (21),(22) and (23) form a scheme that can be applied directly in a lattice-gas simulation. The above scheme is even more efficient if we use *all* particles in the lattice gas as a starting particle, not just one. All the above operations are linear, which means that we do not have to distinguish contributions from different starting particles, we can just simply add the contributions from different tagged particles and use one total probability $P(\mathbf{r}, t)$, which now represents the sum of the 'arriving probability' of all tagged particles. One only has to correct for the fact that different starting particles have different initial velocities. This is taken into account if we weigh the initial value $P(\mathbf{r}, 1)$ by the initial tagged particle velocities, and subsequently leave the initial velocity out of Eq. (21):

$$P(\mathbf{r}, 1) = n_i(\mathbf{r} - \mathbf{c}_i, 0) c_{ix} . \tag{24}$$

The evolution of P is still given by Eq. (22), and the VACF at time t is then

$$\langle v_x(0)v_x(t)\rangle_{tr} = \frac{1}{N}\sum_{\mathbf{r}} P(\mathbf{r};t)\tilde{u}_x(\mathbf{r},t)\ , \tag{25}$$

where N is the total number of particles. Equations (22), (24) and (25) are the basic equations of the MP-method, which can be directly evaluated in a normal lattice-gas simulation, with only a few extra lines of code. Equations (22) and (24) show that the essence of the method is the propagation of the first moment of the single-particle distribution function; in general one can propagate any moment of the distribution function, or any other single-particle property. Unfortunately, this means that the method cannot be applied to calculate for instance the stress-stress correlation function, as the stress tensor is a collective property of the system. Note that the VACF is only averaged over all possible trajectories of all starting particles of one particular initial configuration. The big advantage of this method is that the number of possible trajectories increases with time, hence the statistical accuracy increases. Simulations in two- and three dimensions show that the statistical error decreases approximately as t^{-1}. For longer times, the VACF itself decays as t^{-1} and $t^{-1.5}$ in 2D and 3D respectively, so the signal-to-noise ratio is nearly constant in time. As an example, see Fig. 1. To increase the overall accuracy, additional ensemble averages can be performed. We finally note that the idea of the MP-method has also been applied in very different fields, such as polymer physics [13, 14] and quantum simulations [18].

IV. RESULTS IN TWO, THREE, AND FOUR DIMENSIONS

In this section we briefly review the simulation results obtained for the tagged-particle velocity autocorrelation function in two-, three- and four-dimensional LGCA, with the use of the moment-propagation method. In the simulations we have used various system sizes, and periodic boundary conditions. In all cases correlations were only computed for time intervals less then the shortest time in which any particle could cross the periodic box. This means for instance that for a 3D system of $50 \times 50 \times 50$ lattice sites we only compute the VACF up to 50 time steps, which is in any case less than the acoustic-wave traversal time; this in contrast to corresponding simulations of long-time tails in atomic fluids [15] with the use of molecular dynamics, where

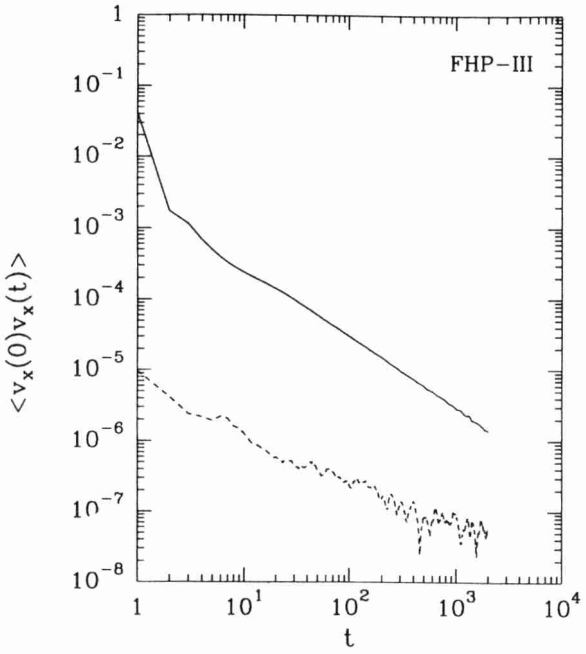

Fig. 1. Typical example of a simulation result for the velocity autocorrelation function of a tagged particle, with the use of the moment-propagation method (solid curve), in a two-dimensional lattice-gas system. The VACF is normalized to one at $t = 0$, and drawn on a log-log scale. Note that the error (dashed curve) decreases with increasing time, which gives an almost constant signal-to-noise ratio. This feature allow us to obtain errors of the order 10^{-8}, which is orders of magnitude lower than can be achieved by brute force (constant error in time). The error was estimated as the standard deviation in the results of 9 independent simulations. The calculation took some 16 hours on a NEC-SX2 (1.1 Gflop) supercomputer.

time intervals up to 5 times the acoustic wave traversal time had to be used. We should mention that for lattice gases the theoretical prediction for the amplitude of the long-time tails is slightly modified [8, 11], and will be a factor $(1 - \rho/b)v_0$ smaller than given in (9), in which v_0 is the volume of a unit cell in the lattice, and ρ is in units of particles per site. We stress that in general the theoretical and simulation results are obtained completely independently and that in the comparison we do not make use of any adjustable parameter.

Typical examples of the VACF obtained in two-, three-, and four dimensional lattice-gas simulations are shown in Figs. 2a-c, on a log-log scale. In these figures we plotted the two extreme theoretical predictions : the dashed line represents the exponential decay that follows from the Boltzmann assumption (no correlations), and is valid for very short times. The solid line represent the prediction for the VACF in the long-time limit as predicted from mode-coupling theory (9). Up to now there has been no full theory to account for the intermediate behavior. As we can see from figure 2a-c, we find good agreement between the measured correlation function (squares) and the corresponding theories, in both time regimes. For a more detailed comparison between the simulation results and mode-coupling theory we refer to [8].

V. EVIDENCE FOR 'FASTER-THAN-T^{-1}' DECAY

In this section we concentrate on the long-time decay of the two-dimensional VACF; the simulation were preformed in essentially the same way as described in the previous section, however, the present calculations were specifically aimed at observation of deviations from t^{-1} decay, and are therefore much longer: we used a system size of 2000 $\times$ 2000 lattice sites, and the correlation time interval of 2000 lattice-gas time steps. All the simulations reported here were carried out in the FHP-III model [9] at densities $\rho = 4.90$, $\rho = 5.25$ and $\rho = 5.60$, in units of average number of particles per site (maximum possible is 7). We did not simulate at any other density, because 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 [16]. The simulations reported here took some 100 hours on a NEC-SX2 supercomputer, and some 50 hours on a single CRAY-YMP processor.

All the simulations results are shown in Fig. 3, where we we plotted the measured VACF multiplied by the time t, for densities $\rho = 4.90$(A),$\rho = 5.25$(B) and $\rho = 5.60$(C). Time is in units of the mean free time t_0, which is determined by the by the initial exponential decay $\sim \exp(-t/t_0)$. Note that we have measured the VACF up to 6000 mean free times, without interference of the periodic images, and with an error of at most 5%. If the VACF deviates from the predictions of lowest-order mode-coupling theory, in which the transport coefficients D and ν take the (time-independent) Boltzmann values,

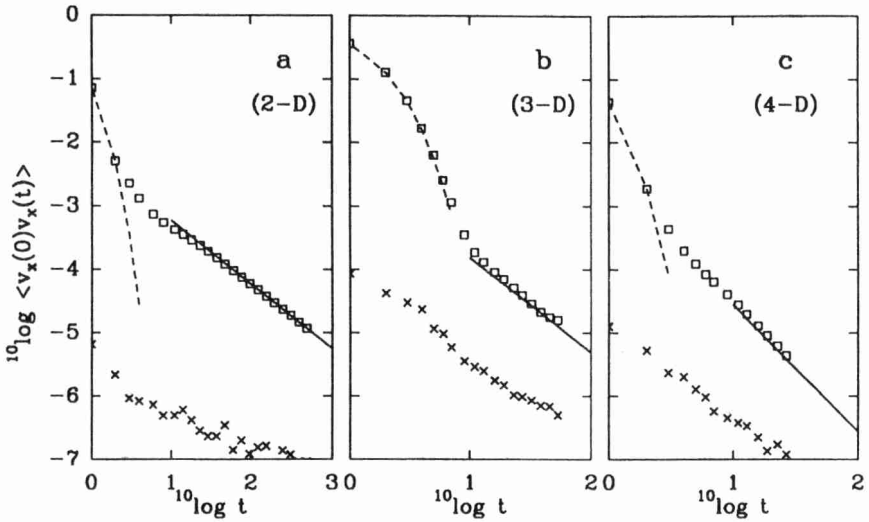

Fig. 2a-c. Normalized velocity autocorrelation function of a tagged particle in a two-,three- and four-dimensional lattice gas fluid. The squares are the data obtained from simulation using the moment propagation method. The crosses represent the statistical error. The dashed lines represent the theoretical predictions in the Boltzmann assumption. The solid lines are the predictions from mode-coupling theory for the long-time decay of the VACF. The specific models are (a) two-dimensional FHP-III model, at density $\rho = 4.9$, (b) three-dimensional FCHC model, at density $\rho = 2.4$, (c) four-dimensional FCHC model, at density $\rho = 12.0$.

the simulation results should differ significantly from the dashed horizontal lines in Fig. 3. 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. 3. at density $\rho = 5.25$ and $\rho = 5.60$. 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 $\rho = 4.90$ (A), where the faster-than-t^{-1} decay is almost unobservable due to the relatively large statistical errors.

Let us next consider a comparison with higher-order mode-coupling theory. In that case, we should use the mode-coupling prediction (10) with

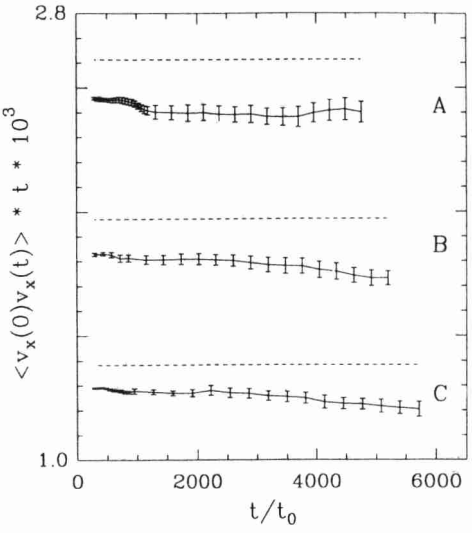

Fig. 3. Velocity autocorrelation function in a two-dimensional lattice gas, multiplied by time, for densities $\rho = 4.90$(A),$\rho = 5.25$(B) and $\rho = 5.60$(C). The solid curves denote the simulation-data (with error bars). The fact that all these curves have, on average, a negative slope is indicative of faster-than-t^{-1} decay. The dashed lines are the predictions of lowest mode-coupling theory (10), using the (known) Boltzmann transport coefficients. t_0 equals 0.38, 0.35 and 0.31 for systems A, B, and C respectively, in units of Δt.

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 (see Eqs. (1) and (4)). In the same way, the time-dependent viscosity can be obtained by integrating the stress-stress correlation function; however, the latter could not be measured in the simulation with sufficient accuracy. Fortunately, we can make a good theoretical estimate. The results for the VACF showed that the deviations from Boltzmann in the decay of the VACF are almost exclusively due to the long-time tails themselves. Lowest-order mode-coupling theory predicts that these long-time tails are of the form t^{-1}, so we estimate that the correction to the Boltzmann viscosity ν_0 is given by

$$\nu(t) = \nu_0 + \int_{t_i}^{t} \frac{c_0}{s} ds \,, \tag{26}$$

where t_i is the time at which the long-time tail sets in; we estimated that this happens after about ten time steps. An expression for the tail amplitude c_0 has been derived by Ernst *et al* [16] for lattice gases. Strictly speaking this is not a comparison with the true self-consistent mode-coupling theory, as we did not use the self-consistent form of the long-time tail in estimating $\nu(t)$. However, for the times studied the differences in $\nu(t)$ are expected to be negligible. Having thus obtained the time-dependent D and ν, we can compare the measured VACF with the higher-order mode-coupling theory. The comparison with the simulation data is shown in Fig. 4 for density $\rho = 5.25$ only; the solid curve with error bars represents the simulation result, the dashed line is the prediction of lowest-order mode-coupling theory (both same as in Fig. 3). The dashed-dotted curve is the prediction of higher-order mode-coupling theory. From this figure we can draw two important conclusions. Firstly, the magnitude of the tail predicted by higher-order mode-coupling theory is in agreement with the simulations. Secondly, the faster-than-t^{-1} decay observed in the simulation is compatible with the decay predicted on basis of knowledge of the time-dependent transport coefficients in (11). We stress that the latter is the basic idea behind self-consistent mode-coupling theory. For the other two densities we found similar agreement [17].

VI. CONCLUSIONS AND DISCUSSION

What we have hoped to demonstrate in this contribution is that, by making use of a new and efficient simulation technique, we have been able to make some qualitatively new observations on the 'long-time tails'; in particular we found that the long-time tail of a particle in a 2D fluid decays faster than t^{-1}. It would be interesting to determine whether the functional form of the observed VACF is compatible with the *full* self-consistent prediction (13). However, in order to observe the latter behavior, the divergent contributions to D and ν should exceed the Boltzmann values D_0 and ν_0. A simple estimation of the time when this is expected to happen goes as follows: we estimate the time-dependent diffusion coefficient by

$$D(t) = D_0 + \int_{t_i}^{t} \frac{b}{s} ds \,,$$

and the time-dependent viscosity $\nu(t)$ by (26). The constant part in the de-

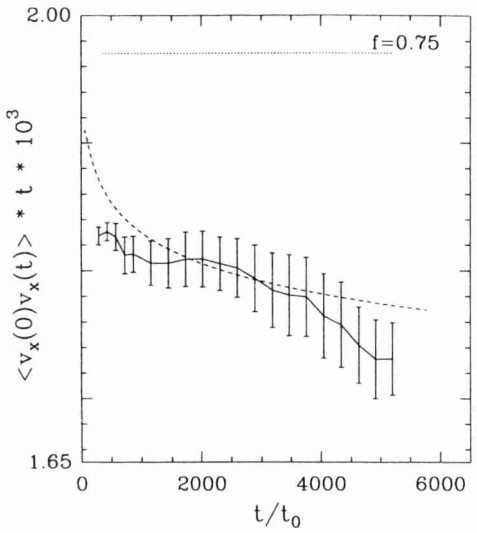

Fig. 4. Velocity autocorrelation function of a tagged particle multiplied by time, for density $f = 5.25$. Solid curve: simulation data (same as Fig. 3); Dashed line: prediction of lowest-order mode-coupling theory; Dashed-dotted curve: prediction of mode-coupling theory with the 'true' time-dependent transport coefficients. The time-dependent diffusion coefficient is obtained from the measured VACF, the time-dependent viscosity is estimated from lowest-order mode-coupling theory.

nominator of (10) is then *equal* to the diverging part if:

$$\ln(t/t_i) = \frac{D_0 + \nu_0}{b + c_0}$$

If we substitute the values for one particular density, for instance $\rho = 5.6$, we find that $\ln(t/t_i) \approx 41$, or

$$t/t_i \approx 10^{18}$$

where t_i is of the order of 10. Again we only made use of the lowest-order prediction for the time-dependent transport coefficients. However, higher-order self-consistent predictions would give a slower divergence of the transport coefficients, which would yield even larger values for t. So we estimate that the full self-consistent prediction (13) can, at its earliest, be observed 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 lattice-gas models.

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