Fast Algorithms for Slow Processes in Lattice-Gas Cellular Automata

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ABSTRACT

The accurate numerical study of long-time tails in time-correlation functions requires large amounts of computer time. In some cases, the necessary calculations are too time-consuming to be carried out even on present-day (super)computers. However, if we consider 'lattice-gas' versions of the same problems, then it turns out that for certain problems, a speed-up of 6-10 orders of magnitude can be achieved using a very simple algorithm. As a result, we can now test theoretical (mode-coupling) predictions for long-time tails with unprecedented accuracy.

1 Introduction

The algorithms that we use in Monte Carlo and Molecular Dynamics simulations of atomic or molecular systems have not changed fundamentally since they were first introduced in the fifties and early sixties [1]. This fact clearly illustrates something that hardly needs illustrating, namely that the 'founding fathers' of computer simulation were exceptional scientists. At the same time, this observation is a bit depressing. Even after 35 years of rapid growth in the number of applications of computer simulation to problems in physics and chemistry, the main increase in computing speed is still simply due to the increased speed of computers. In other words: there are no short-cuts in the simulation of atomic and molecular fluids. The number of floating-point operations in a typical simulation may be somewhat larger than strictly necessary, but not by orders of magnitude.

This is indeed depressing news because even with today's supercomputers there are numerous properties of atomic or (macro)molecular systems that defy numerical simulation. As an example, I shall consider the numerical study of the velocity autocorrelation-function of a tagged particle in a fluid.

1.1 Long-Time Tails

In the history of the kinetic theory of fluids, 1969-1970 was a crucial year. In that year Alder and Wainwright [2] published a paper in which they demonstrated the breakdown of the 'Molecular Chaos' assumption. The Molecular Chaos assumption, originally introduced by Boltzmann as the 'Stoßzahlansatz', states that the collisions experienced by a molecule in a fluid are uncorrelated. One consequence of this assumption is that the velocity autocorrelation function (VACF) of a tagged particle in fluid should decay exponentially. What Alder and Wainwright found is that the VACF of a particle in a moderately dense fluid of hard spheres or hard disks

does not decay exponentially but algebraically. These algebraic long-time tails are the consequence of coupling between particle diffusion and shear modes in the fluid.

The Alder-Wainwright simulations caused a complete overhaul of the kinetic theory of dense fluids. The subsequent theoretical analyses of algebraic long-time tails were either based on an extension of kinetic theory [3] or on mode-coupling theory [4]. For a review, see [5]. In the mode-coupling theory by Ernst, Hauge and van Leeuwen [4], it is assumed that the long-time tail is the consequence of coupling between particle diffusion and shear modes in the fluid. To a first approximation the leading term in the long-time tail of the velocity ACF is given by:

$$\langle v_x(0)v_x(t)\rangle \approx \frac{D-1}{D}\frac{1}{\rho[4\pi(D_0+\nu_0)t]^{D/2}} \equiv \frac{d_0}{t^{D/2}}\,, \tag{1}$$

where ρ is the number density, D_0 the 'bare' self-diffusion constant, ν_0 the kinematic viscosity and D the dimensionality. These 'bare' transport coefficients are obtained by computing the diffusion constant and the kinematic viscosity using Boltzmann's molecular chaos assumption, i.e. ignoring any contribution of the long-time tails themselves. In a self-consistent mode-coupling theory, the transport coefficients that enter into the expression for the long-time tails are computed using information about the long-time tails themselves. I shall return to this point below.

Following this theoretical work, simulations were performed by Levesque and Ashurst [6] and, most extensively, by Erpenbeck and Wood [7,8] with the aim to verify the validity of eqn. 1. For three-dimensional fluids these simulations are extremely expensive because very long simulations on very large systems must be performed. Even so, the system sizes studied in the simulations of Erpenbeck and Wood were such that it was essential to apply finite-size corrections to the corresponding mode-coupling theory before a meaningful comparison with the simulations could be made. Following such an approach, Erpenbeck and Wood found agreement between their simulation results for the VACF and a finite-size mode-coupling theory for a number of different densities. Nevertheless the statistical accuracy of their data was such that it was not meaningful to verify either the value of the exponent of the algebraic tail or the functional form of the density-dependent tail coefficient independently.

In the case of two-dimensional fluids there is another problem. Ever since the discovery of hydrodynamic tails, it has been realized that a consistent description of mode-coupling effects in a two-dimensional fluid would result in a long-time tail that decays faster than t^{-1}, because in $2D$ the self-diffusion constant D diverges. Moreover, the viscosity, which is proportional to the integral of the stress autocorrelation function, is also expected to diverge. Alder and collaborators resolved this difficulty by introducing time-dependent transport coefficients [9]. In this way, the hydrodynamic description could be made self-consistent. This self-consistent mode-coupling theory predicts that the decay of the VACF is slightly faster than t^{-1}, namely

$$\langle v_x(0)v_x(t)\rangle \sim \frac{1}{t\sqrt{\ln t}}. \tag{2}$$

Unfortunately, this correction to the t^{-1}-decay was too weak to be observed in Alder's simulations or, for that matter, in any subsequent simulation of hard disks.

Of course, one may argue that long-time tails themselves are almost impossible to detect experimentally and that this holds *a fortiori* for small corrections to these tails in two-dimensional fluids. However, the concepts that underly the self-consistent mode-coupling picture are used in many branches of physics. In that context, a direct test of this proto-typical self-consistent mode-coupling effect is of considerable importance.

In summary: the direct observation by 'conventional' computer simulation of corrections to the t^{-1} decay of the velocity ACF in a two-dimensional fluid is a problem that requires much more computing power than is presently available. Below, we shall see that with 'non-conventional' simulation techniques, the effect can, in fact, be observed.

2 A Simplified Model

The problem of transport in an atomic fluid can be cast in a simplified, a space-time discretized form if we use Lattice Gas Cellular Automata (LGCA's) to model a simple fluid.

In LGCA's, the 'atoms' are constrained to move along the bonds joining the lattice sites. No two particles can move along the same bond in the same direction. The state of the lattice is completely specified by indicating which links are occupied and which are empty. This implies that lattice-gas particles are indistinguishable.

The time evolution of the system is governed by the following rules. :

1. Propagation: all particles move in one time step (for convenience we choose $\Delta t = 1$) from their initial lattice position (say $\mathbf{X}$) to a new position ($\mathbf{X}' = \mathbf{X} + \mathbf{c}_\alpha$; where $\mathbf{c}_\alpha$ is the velocity of species α). The velocities $\mathbf{c}_\alpha$ are such that at the end of the propagation steps all particles are once more positioned at lattice sites.

2. Collision: the particles at all sites on the lattice undergo a collision that conserves the total number of particles and the total momentum at each site. These local collision rules may, or may not, be deterministic.

Provided that the lattice has a sufficiently high symmetry (e.g. triangular in 2 dimensions) and the collision rules are sufficiently isotropic (for a discussion, see [11]), it can be shown that the equation that governs the time evolution of the 'flow field' of such a lattice gas becomes equivalent to the Navier-Stokes equation for an incompressible fluid in the limit that the flow velocity is much less than the particle velocity, and all spatial variations in the system occur on a scale that is large compared to the mean free path of the lattice gas particles. In this respect LGCA's model atomic fluids. In particular, we should expect to observe long-time 'hydrodynamic' tails in the velocity ACF of a tagged particle in a lattice gas.

2.1 Moment Propagation

What is it that makes the calculation of long-time tails so difficult? The problem is that we must sample a particular quantity (the product of the tagged-particle velocity at two different times) over many trajectories. However, in a conventional

simulation we generate much more information than we need: a Molecular Dynamics simulation will provide information about the complete history of all particles in the system, not just about one particular time-correlation function. Below, I shall indicate that it is possible to gain many orders of magnitude in computing speed, if we are willing the sacrifice such detailed, but usually irrelevant, information. The 'moment-propagation' method [12-14] allows us to compute certain averages of the n-body distribution function directly, without attempting to compute the function itself. This approach results in a very appreciable gain in computing speed: for instance, in the case of velocity autocorrelation functions in lattice gases, the method has resulted in a speed-up that varies between 10^6 and 10^{10} [12,13].

In order to explain the method, it is most convenient to consider one tagged particle, say a 'blue' particle, in a system of, otherwise identical, 'red' particles. If the 'blue' particle collides with one or more red particles, it is no longer possible to tell which one of the outgoing particles is the 'blue' particle. We make use of this fact by defining stochastic collision rules such that the 'blue' particle has equal probability to end up in any of the outgoing states. Of course, the actual 'colour-blind' dynamics of the lattice gas is not affected by the way we tag particles. Hence there are many possible trajectories of the tagged particle that are all compatible with the same sequence of 'un-coloured' states of the lattice-gas. All these tagged-particle trajectories will, in general, have different statistical weight. For any trajectory, this weight is simply the product of the successive scattering probabilities of the tagged particle. The average of $v_x(0)v_x(t)$ of the 'blue' particle is the weighted sum over all paths.

It should be stressed that, in order to compute the velocity ACF, we do not need to know which specific path a particle has followed. On the contrary, we only need to compute the probability that the 'blue' particle is at site $\mathbf{r}$ at time t multiplied by its velocity at time $t = 0$. All particles with trajectories leading to $\mathbf{r}$ at time t will have the same average post-collisional $v_x(t)$. The probability to find a particle at $\mathbf{r}$ at time t is simply the sum of the probabilities of all trajectories ending at this site. This quantity can easily be computed recursively: if we know the total probability $P(\mathbf{r}', t-1)$ of all trajectories that terminate at any of the positions $\mathbf{r}'$ *neighboring* to $\mathbf{r}$ at time $t-1$, we compute the probability to be at $\mathbf{r}$ at time t by simply propagating these $P(\mathbf{r}', t-1)$ to $\mathbf{r}$, multiplied with the probability that there is, in fact, a tagged particle moving from $\mathbf{r}'$ to $\mathbf{r}$ in this time interval. The propagated probabilities are then simply added to yield the desired probability that the tagged particle will be at $\mathbf{r}$ at time t. In practice, what we compute is not the probability $P(\mathbf{r}, t)$ to find any tagged particle at $\mathbf{r}$ at time t, but it is this probability *multiplied* by the initial velocity of the tagged particle.

It is important to note that we can choose with equal probability *any* particle on the lattice as the tagged particle. The contributions for all different starting positions are automatically added in this algorithm. In summary, the correlation function $\langle v_x(0)v_x(t)\rangle$ is evaluated as the weighted sum over all sites of the lattice and all possible tagged particle trajectories of the product of the post-collisional velocity $v_x(t)$ and the initial velocity of the tagged particle $v_x(0)$. More generally, rather than propagate the initial velocity of the tagged particle along all possible trajectories, we can propagate any moment of the single-particle distribution function.

The method is conveniently summarized by the following equations : Let us define the number of particles N, as

$$N = \sum_{\mathbf{r}} \sum_{i} s_i(\mathbf{r}, 0) ,$$

where $s_i(\mathbf{r})$ is one if there is a particle at site $\mathbf{r}$ moving with x-velocity c_{ix} in the direction of bond i, and zero otherwise. The average post-collisional velocity of a tagged particle at site $\mathbf{r}$ is:

$$\bar{v}_x(\mathbf{r}, t) = \sum_{i} \left[\frac{s_i(\mathbf{r}, t)}{\sum_j s_j(\mathbf{r}, t)} c_{ix} \right] .$$

The sum over all tagged particles of the probability to find one individual tagged particle at site $\mathbf{r}$ weighted with its initial velocity is denoted by $W(\mathbf{r}, t)$, and evolves in time as :

$$W(\mathbf{r}, t+1) = \sum_{i} \left[W(\mathbf{r} - \mathbf{c}_i, t) \frac{s_i(\mathbf{r} - \mathbf{c}_i, t)}{\sum_j s_j(\mathbf{r} - \mathbf{c}_i, t)} \right] \qquad \forall \mathbf{r} ,$$

where the initial value $W(\mathbf{r}, 1)$ is given by

$$W(\mathbf{r}, 1) = \sum_{i} s_i(\mathbf{r} - \mathbf{c}_i, 0) c_{ix} .$$

The VACF at time t is now simply given by

$$\langle v_x(0) v_x(t) \rangle = \frac{1}{N} \sum_{\mathbf{r}} W(\mathbf{r}, t) \bar{v}_x(\mathbf{r}, t) .$$

Note that the averaging is over all possible paths of one tagged particle and in turn over all possible particles. After t timesteps, the number of possible paths of one particle is of the order ρ^t, where ρ is the average number of particles per site. As a result the averaging is over approximately $N\rho^t$ different events, although not uncorrelated. Nevertheless, this greatly improves the statistics and, most importantly, the absolute error decreases with increasing t. This feature makes the method extremely useful for calculating the VACF for longer times.

3 Results

In this section I show an example that illustrates the power of the moment propagation method. This example concerns the observation of self-consistent mode-coupling effects in hydrodynamic long-time tails. The example is merely intended as an illustration. For a detailed discussion, the reader is referred to the relevant publication [15].

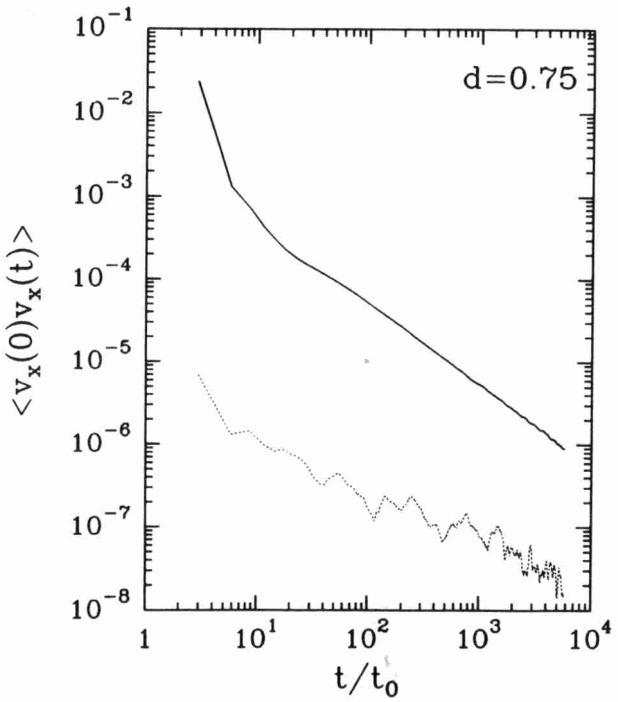

Figure 1: Velocity ACF of a tagged particle in a two-dimensional lattice-gas cellular automaton (FHP-III model, see [10]). The density d denotes the fractional filling of the lattice. Time is expressed in units of the 'mean-free time' t_0. t_0 is the time that characterizes the initial ('Boltzmann') decay of the VACF. At this density, t_0 equals 0.346 time steps of the lattice gas. The best simulations of the velocity ACF of hard disks extend to some 10^2 mean-free times.

The velocity ACF is indicated by a drawn curve, while the estimated statistical error is shown as a dotted curve. Note that the signal-to-noise ratio hardly deteriorates with increasing time. Note also that the statistical noise at long times is of the order of 2×10^{-8} which is 5 orders of magnitude better than the best 'conventional' result for the same model system.

3.1 Long-Time Tails in Lattice Gases

Lattice-gas cellular automata were first introduced as a model for hydrodynamic fluids [16] as a computationally 'cheap' model for a simple fluid. The number of collisions per CPU-second in a lattice gas is at least 4 order-of-magnitude larger than in a hard-sphere fluid. It was therefore hoped that the direct simulation of the tagged-particle motion in a LGCA would yield very accurate data on the algebraic decay of the VACF. In fact, very extensive simulations on two-dimensional LGCA's were performed by several groups [17,18] with precisely this aim. But the results were disappointing. In fact, the statistics on the long-time tails were poor (typically, an accuracy of 0.3% was the best that could be achieved) and the tails were almost completely swamped in noise. We have applied the moment-propagation method to a model for a lattice-gas on a triangular lattice (to be precise, the 'FHP-III'-

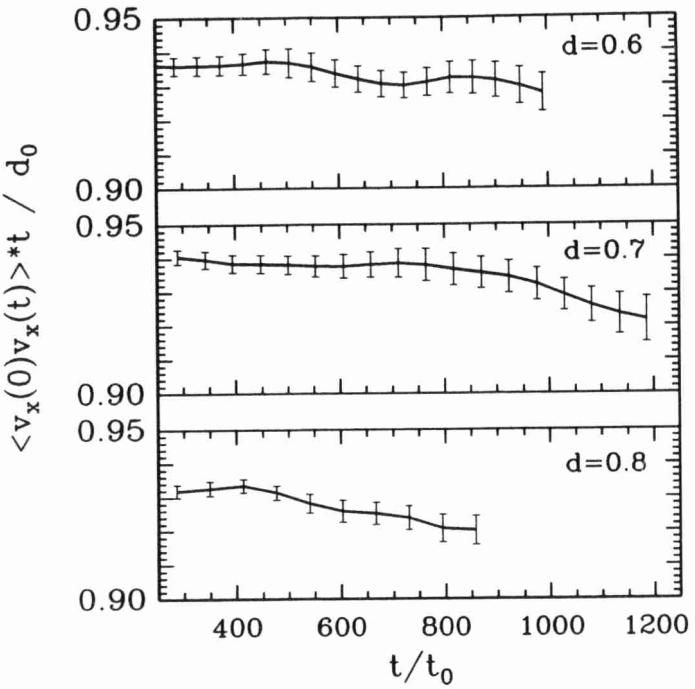

Figure 2: This figure shows the long-time tail of a tagged particle in a two-dimensional lattice-gas cellular automaton, divided by the tail predicted by lowest order mode coupling theory. In this theory, the expression for the hydrodynamic tail is of the form d_0/t (see eqn. 1). The explicit expression for d_0 is given in [19]. The density (i.e. the fractional filling of the lattice) is denoted by d. The time is expressed in units t_0 (see Fig. 1). For d =0.6,0.7 and 0.8, t_0 equals 0.454, 0.379 and 0.315, respectively.

model described in ref [10]). Use of the moment-propagation method results in a very drastic reduction of the statistical noise by a factor that can be as much as 10^5 for long times (see Fig. 1). This noise reduction would correspond to a gain of 10^{10} in computing speed. Admittedly, we have not compared the two techniques on the same computer, but this is not expected to change the picture by more than a factor 10^3 (and probably less). Note that Fig. 1 shows a very clear non-exponential decay at long times. We can now compare the results of such a simulation with the predictions of lowest order mode-coupling theory (see [19]). Such a comparison is shown in Fig. 2. The figure clearly shows that the observed tail falls below the predictions of lowest-order mode coupling theory at all densities studied. It should be noted however that the truly asymptotic regime described by eqn. 2 only dominates at times of the order of 10^{20} time-steps. This in inaccessible at present, and almost certainly, also in the future. For more details, the reader is referred to [15].

If the simulation results were adequately described by lowest-order mode coupling theory, all curves would be constant at a value of 1.0 . However, the figure

shows that the velocity ACF is in all cases *lower* than the value predicted by the first-order mode-coupling theory. Moreover, all curves show, on average, a weak, negative slope. Both effects are in agreement with the predictions of self-consistent mode-coupling theory.

4 Conclusion

In this paper I have shown how, sometimes, a change of algorithm can make a difference. I have discussed a novel numerical technique to study time-correlations in 'sub-molecular' fluids, and I have shown how using this algorithm, we can observe interesting phenomena that were, thus far, inaccessible.

However, the application of the 'moment propagation' algorithm is not limited to the study of time correlations. The same scheme has been extended to study the structure and phase-behavior of 'supra-molecular' fluids. This opens the way to the numerical study of a class of problems that did, thus far, defy simulation. For a discussion of this aspect of the algorithm, the reader is referred to [20,21].

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