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The super long-time decay of velocity fluctuations in a two-dimensional fluid

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Abstract

We have calculated the velocity autocorrelation function for a tracer particle in a model two-dimensional fluid. The fluid was represented by a lattice Boltzmann equation with imposed fluctuations. By choosing a low Boltzmann diffusion coefficient for the tracer, the diverging contribution to the diffusion coefficient can be made to exceed the Boltzmann value even at short times. We were thus able to find evidence for the renormalized, or 'super long-time', decay of the VACF in a two-dimensional fluid. We find quantitative evidence for the $1/t\sqrt{\ln(t)}$ decay predicted by theory.

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1. 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 showed that velocity fluctuations do not decay exponentially, as had previously been assumed, but algebraically. From simple hydrodynamic arguments they showed that at long times the velocity autocorrelation function (VACF) decayed as $t^{-d/2}$, in a d -dimensional fluid. In terms of the α component of the particle velocity, v_α , the velocity autocorrelation function, $\phi(t)$, is defined as follows:

$$\phi(t) = \langle v_\alpha(0)v_\alpha(t) \rangle, \quad (1)$$

and it is related to the diffusion coefficient, D , by the Green–Kubo relation

$$D = \int_0^\infty \langle v_\alpha(0)v_\alpha(t) \rangle dt. \quad (2)$$

So, the presence of 'long-time tails' has important consequences for the calculation of transport coefficients. This is most dramatic for a two-dimensional fluid where the Alder and Wainwright result, $\phi(t) \sim t^{-1}$, implies that the diffusion coefficient does not exist. The integral in Eq. (2) diverges.

Following in the spirit of the original work, mode-coupling [2] and kinetic theories [3] were developed to provide a theoretical framework for the description of these long-time tails. Both classes of theory reproduce the original Alder and Wainwright result, but also yield expressions for the constant of proportionality in the long time decay. The lowest order mode-coupling prediction for the VACF in two dimensions is [2]

$$\phi(t) = \frac{m\phi(0)}{8\pi\rho(D + \nu)t}, \quad (3)$$

where m is the particle mass, ρ the fluid density and ν the kinematic viscosity of the fluid. This analysis assumes that D and ν are constant. Of course this cannot be true for the diffusion coefficient. If D is constant then Eq. (3) also says that D is diverging as $\ln(t)$, so it is inconsistent. The same is actually true for the viscosity, which is related to the time integral of the stress autocorrelation function (SACF). This is also predicted to diverge logarithmically in two dimensions [2]. Wainwright et al. [4] suggested that a consistent picture could be obtained by considering time dependent transport coefficients $D(t)$ and $\nu(t)$, defined as the time integrals of the VACF and SACF, respectively. By replacing D and ν in Eq. (3) with their time dependent counterparts, one has

$$\phi(t) = \frac{m\phi(0)}{8\pi\rho(D(t) + \nu(t))t}. \quad (4)$$

A consistent picture is obtained by finding long time behaviour for $D(t)$ and $\nu(t)$ which, when substituted into Eq. (4) and the equivalent expression for the SACF, implies the correct decay for the VACF and SACF. It turns out that the VACF decays somewhat faster as a result of this renormalization, i.e. as $1/t\sqrt{\ln(t)}$ instead of $1/t$. The formal expression is

$$\phi(t) = \sqrt{\frac{m\phi(0)}{16\pi\rho}} \left(\frac{1}{t\sqrt{\ln(t)}} \right). \quad (5)$$

This result can also be shown explicitly by summing all mode-coupling contributions to the long-time tail [5].

Of course at short times ($t < \tau_B$, where τ_B is a few mean collision times), the algebraic tail is irrelevant. The motion of a tagged particle can be represented by the Boltzmann equation (which ignores correlated motions). The Boltzmann value for the diffusion coefficient does not diverge – the theory does not contain the long time effects which cause the divergence. The true diffusion coefficient can be thought of as consisting of two components. A short time Boltzmann contribution, D_B , which converges on a time scale τ_B , and a long time non-Boltzmann contribution, $D_{NB}(t)$, which we define as $D_{NB}(t) = D(t) - D_B$. It is this non-Boltzmann contribution which diverges on a time scale $t \gg \tau_B$. If we ignore the effect of the SACF then, based only on the relative

magnitudes of $D_{NB}(t)$ and D_B , we can identify two possible pre-asymptotic regimes in the long time ($t \gg \tau_B$) decay of the VACF;

- (1) $D_B \gg D_{NB}(t)$ – where renormalization effects are negligible and the decay of the VACF should be adequately approximated by Eq. (3), with D_B replacing D in the denominator.
- (2) $D_B \approx D_{NB}(t)$ – where renormalization effects begin to influence the VACF and Eq. (4) should adequately describe the decay.

However, at sufficiently long times the diverging contribution must exceed the Boltzmann contribution and when $D_{NB} \gg D_B$ the truly asymptotic decay (Eq. (5)) should occur. In this discussion we have only considered the divergence of $D(t)$. In fact the result for the VACF given in Eq. (5) is obtained if one does, or does not, consider the divergence of $\nu(t)$. We do, however, have to satisfy the additional condition that the time dependent diffusion coefficient and not the time dependent viscosity is dominating the decay of the VACF (see Eq. (4)). So we must have $D(t) \gg \nu(t)$ for Eq. (5) to be valid.

The most extensive tests of theoretical predictions for $\phi(t)$ in a two-dimensional fluid were performed on a simple model fluid, namely a lattice gas cellular automaton of the type introduced by Frisch, Hasslacher and Pomeau [6]. In such a system the fluid particles are constrained to move on a lattice. By making use of this fact Frenkel and Ernst [7] developed a technique, known as moment propagation, which allowed the VACF to be calculated to an accuracy four orders of magnitude better than had hitherto been possible. Using this technique van der Hoef and Frenkel [8] observed the first pre-asymptotic regime (regime (1)). Following more extensive simulations [9] they also observed the onset of the second pre-asymptotic regime (regime (2)). The relative weakness of the long-time tail in this lattice gas led van der Hoef and Frenkel to conclude that the true asymptotic decay would only be observable after about 10^{20} collision times [10]. Subsequently, Leegwater [11] argued that it might take even hundreds of orders of magnitude longer than that, clearly illustrating why the $1/t\sqrt{\ln(t)}$ decay is sometimes referred to as the 'super' long-time tail. In this article we describe calculations of the VACF in a model fluid closely related to a lattice gas – the lattice Boltzmann fluid. By using the greater freedom this model gives us to vary the magnitudes of the Boltzmann and non-Boltzmann contributions to the VACF, we were able to find evidence, in a computer simulation, for the super long-time behaviour of the VACF in two dimensions.

2. Description of the model

In order to explain the lattice Boltzmann model that we have used [12–14], it is useful to briefly describe lattice gases. A lattice gas cellular automaton is an idealized model fluid in which particles move with a discrete set of velocities, c_i , on a lattice. The state of the fluid at any (discrete) time is defined by a set of Boolean variables, $s_i(\mathbf{r}, t)$, which describe whether a particle with velocity c_i is, or is not, located at node \mathbf{r} at time t (an exclusion rule applies so states cannot be multiply occupied). The time evolution

of the lattice-gas comes in two parts; propagation, during which every particle moves along its link to the next lattice site, and collision, where at each lattice site particles can change velocities. As long as the collision step satisfies certain restrictions (it must conserve particle number, momentum, and retain the symmetry of the lattice) then the lattice gas behaves in many respects like a Newtonian fluid. Because the lattice gas contains the hydrodynamics of a real fluid, and theory tells us that it is the slow decay of these hydrodynamic fields that determines the long time decay of the correlation functions, a tagged lattice gas particle should behave in a similar manner to a tagged particle in a real fluid.

The lattice Boltzmann model is a pre-averaged version of a lattice gas. The state of the fluid is specified by the average number of particles at each link, $\langle s_i(\mathbf{r}, t) \rangle$, which we denote $n_i(\mathbf{r}, t)$. The hydrodynamic fields, mass density ρ , momentum density \mathbf{j} and momentum flux density Π are simple moments of the velocity distribution,

$$\rho = \sum_i n_i, \quad \mathbf{j} = \sum_i n_i \mathbf{c}_i, \quad \Pi = \sum_i n_i \mathbf{c}_i \mathbf{c}_i. \quad (6)$$

Like a lattice gas, the system evolves subject to propagation and collision. In going to the lattice Boltzmann equation we make use of 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 subsequent collision partners (although, as we shall see later, these correlations can be recovered by introducing fluctuations into the system).

The time evolution of the distribution functions n_i is described by the discretized analogue of the Boltzmann equation [15],

$$n_i(\mathbf{r} + \mathbf{c}_i, t + 1) = n_i(\mathbf{r}, t) + \Delta_i(\mathbf{r}, t), \quad (7)$$

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 . The collision operator Δ_i is subject to similar restrictions as those imposed for a lattice gas – a complete description is given by Ladd [16]. However, the main effect of the collision operator is to (partially) relax the shear stress at every lattice site. The rate of stress relaxation is related to the kinematic viscosity ν [16]. The lattice used in this work is the four-dimensional Face-Centered Hyper Cubic (FCHC) lattice projected onto two dimensions. This FCHC model is used because square lattices do not have a high enough symmetry to ensure that the hydrodynamic transport coefficients are isotropic.

The analogy between lattice gases and the lattice Boltzmann equation extends to the moment propagation method. The moment propagation method has been described in detail elsewhere [7], but briefly it proceeds as follows. In a lattice gas the probability that a particle moves with a velocity \mathbf{c}_i after a collision is given by $s_i(\mathbf{r}, t)/\rho(\mathbf{r}, t)$ – for the Boltzmann equation it is $n_i(\mathbf{r}, t)/\rho(\mathbf{r}, t)$. The idea is to make use of this fact and propagate in time a quantity $P(\mathbf{r}, t)$, defined as the sum of the probabilities of all trajectories which end at node \mathbf{r} at time t , weighted by the α component of their initial velocities. After one time step we have

$$P(\mathbf{r}, 1) = \sum_i n_i(\mathbf{r} - \mathbf{c}_i, 0) c_{i\alpha}. \quad (8)$$

The time evolution of $P(\mathbf{r}, t)$ is as follows:

$$P(\mathbf{r}, t+1) = \sum_i \frac{n_i(\mathbf{r} - \mathbf{c}_i, t) P(\mathbf{r} - \mathbf{c}_i, t)}{\rho(\mathbf{r} - \mathbf{c}_i, t)} \quad (9)$$

and the VACF is obtained by multiplying $P(\mathbf{r}, t)$ by the probability that a particle at node \mathbf{r} is currently moving with a velocity component $c_{i\alpha}$. Averaging over all sites this gives

$$\langle c_{i\alpha}(0) c_{i\alpha}(t) \rangle = \frac{1}{N} \sum_{\mathbf{r}} P(\mathbf{r}, t) u_{\alpha}(\mathbf{r}, t), \quad (10)$$

where N is the total number of particles in the system and $u_{\alpha}(\mathbf{r}, t)$ is the local fluid velocity ($= j_{\alpha}(\mathbf{r}, t) / \rho(\mathbf{r}, t)$). For the 'pure' lattice Boltzmann model (as described above) this is not very interesting because, for a fluid at rest, u_{α} is zero at every lattice site and all the terms which we can calculate from the moment propagation equations, i.e. $\phi(t \geq 1)$, are also zero. This expresses the fact that, having made the molecular chaos assumption, the motions of the particles become uncorrelated as soon as they have collided. The Boltzmann collision time for the system, τ_B , is therefore just one time step. The VACF is only non-zero before any collisions occur, i.e. for $t < 1$. The value at $t = 0$ is given by

$$\langle c_{i\alpha}(0) c_{i\alpha}(0) \rangle = \frac{1}{N} \sum_{\mathbf{r}} \sum_i n_i(\mathbf{r}, 0) c_{i\alpha} c_{i\alpha} = \frac{1}{\rho_0} \langle \Pi_{\alpha\alpha}(\mathbf{r}, 0) \rangle, \quad (11)$$

where ρ_0 is the average density. If, however, we consider the 'fluctuating' Boltzmann equation [17], as opposed to the pure Boltzmann equation, then things are different. In the Boltzmann equation the spontaneous fluctuations present in a real fluid are killed off by the pre-averaging. In the fluctuating Boltzmann equation they are reintroduced in the form of imposed random fluctuations in the stress tensor. These stress fluctuations set up microscopic velocity fields in the fluid, so $u_{\alpha}(\mathbf{r}, t)$ will no longer be zero and the VACF for times greater than zero will also be non-zero. This 'non-Boltzmann' contribution to the VACF should decay in the correct hydrodynamic manner, i.e. $\phi(t) \sim t^{-1}$, and in turn give rise to the theoretically predicted renormalization of the VACF. The remaining problem concerns the relative magnitudes of the Boltzmann contribution and non-Boltzmann (diverging) contribution to the VACF. For the lattice Boltzmann model which we have used,

$$\langle \Pi(\mathbf{r}) \rangle = c_s^2 \langle \rho(\mathbf{r}) \rangle + \langle \rho(\mathbf{r}, t) u_{\alpha}(\mathbf{r}, t)^2 \rangle, \quad (12)$$

with the speed of sound, c_s , equal to $1/\sqrt{2}$ (all quantities are given in lattice units where the particle mass, time-step and lattice spacing are equal to unity). If we assume that $\rho(\mathbf{r}, t) \cong \rho_0$ (a good approximation at low Mach numbers) this implies that

$$\langle c_{i\alpha}(0) c_{i\alpha}(0) \rangle = \frac{1}{2} + \langle u_{\alpha}^2 \rangle. \quad (13)$$

On the other hand, the first non-Boltzmann term, $\phi(1)$, is equal to $\langle u_\alpha^2 \rangle$. Since the local fluid velocity, $u(\mathbf{r}, t)$, is constrained to be significantly less than the speed of sound, the first non-Boltzmann term is much smaller than the Boltzmann term – we would have to wait a very long time to reach the fully renormalized region of the VACF (the same problem that occurs if one attempts the calculation in a lattice gas). To circumvent this problem we calculated the VACF of a passive tracer which we introduced into the system. The tracer particles are identical to the lattice Boltzmann fluid except they have a probability $\Delta/\rho(\mathbf{r}, t)$ of remaining at the same lattice site. The probability that a tracer particle moves with a velocity component $c_{i\alpha}$ after a collision is $(n_i(\mathbf{r}, t) - \Delta/b)/\rho(\mathbf{r}, t)$, where b is the number of velocities (b is equal to 24 for the FCHC lattice used here). To calculate the VACF of the tracer we replace $n_i(\mathbf{r}, t)$ in Eq. (8) with $n_i(\mathbf{r}, t) - \Delta/b$. The propagation equation (Eq. (9)) is modified to allow for tracer particles staying at the same site, and so becomes

$$P(\mathbf{r}, t + 1) = \sum_i \frac{(n_i(\mathbf{r} - \mathbf{c}_i, t) - \Delta/b)P(\mathbf{r} - \mathbf{c}_i, t)}{\rho(\mathbf{r} - \mathbf{c}_i, t)} + \Delta \frac{P(\mathbf{r}, t)}{\rho(\mathbf{r}, t)}. \quad (14)$$

Eq. (10) can remain unchanged because the Δ terms cancel when we sum over i . The initial value of the VACF for the tracer is

$$\phi(0) = \frac{1}{N} \sum_{\mathbf{r}} \sum_i (n_i(\mathbf{r}) - \Delta/b) c_{i\alpha} c_{i\alpha}, \quad (15)$$

which for our model gives $\phi(0) = 1/2 - \Delta/2\rho_0 + \langle u_\alpha^2 \rangle$ (for $\rho(\mathbf{r}, t) \cong \rho_0$). However, $\phi(1)$ (the first non-Boltzmann term) is still equal to $\langle u_\alpha^2 \rangle$. So, by increasing the parameter Δ we can reduce the magnitude of $\phi(0)$ relative to $\phi(1)$.

Because of the apparent contradiction of looking for 'non-Boltzmann' behaviour in the Boltzmann equation we will recap what we have done. Starting from an atomistic model fluid, a lattice gas, we ensemble average ignoring correlated particle motions. We then have a Boltzmann fluid which contains all the hydrodynamics of a real fluid, but has no spontaneous fluctuations and, on timescales longer than a collision time, no correlations. In a real fluid Kinetic theory tells us that these correlations give rise to the slow *hydrodynamic* decay of fluctuations. In the Boltzmann fluid – which *contains* all the hydrodynamics, imposed fluctuations will decay in essentially the same way as in a real fluid (see for instance Ref. [18]). So, by introducing fluctuations into the Boltzmann fluid we recover the lost correlations. For a tagged particle in this system we have a short time contribution to the VACF arising from ballistic motion between collisions. This is present with or without the fluctuations and so represents the Boltzmann contribution to the VACF. We also have a second contribution which arises from the slow decay of velocity fluctuations. This is *only* present in the fluctuating Boltzmann equation and is the non-Boltzmann contribution to the VACF. Finally, by adding a passive tracer into the system we can, by varying the Boltzmann diffusion coefficient of the tracer, vary the relative magnitudes of the two contributions. We can thus hope to arrange for the Boltzmann and non-Boltzmann contributions to the VACF

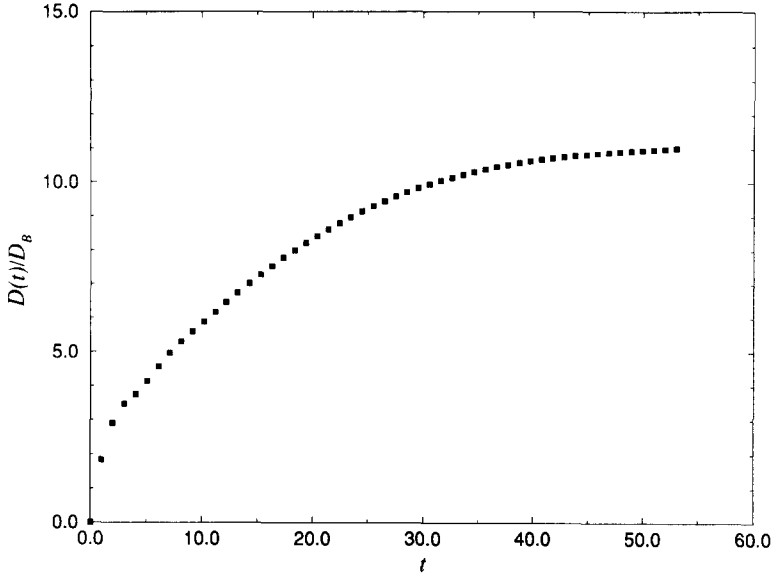


Fig. 1. The time dependent diffusion coefficient, $D(t)$, normalized by its Boltzmann value, D_B , and plotted as a function of time.

to become similar in magnitude at times sufficiently short for us to observe super long time diffusion in a simulation.

3. Results

We used a fluctuating Boltzmann fluid of viscosity 1.0×10^{-3} , density 24 and mean square velocity 3.76×10^{-3} . The tracer had a Boltzmann diffusion coefficient of 5.94×10^{-3} . Notice that the Boltzmann viscosity was significantly smaller than the Boltzmann diffusion coefficient. The aim of this was to ensure that the time dependent viscosity was less than the time dependent diffusion coefficient. Although the difference was not as high as we would have liked, it seems reasonable to assume that, with these parameters, $D(t)$ and not $\nu(t)$ will be playing the dominant role in the decay of the VACF. As we noted earlier, if this were not the case, Eq. (5) would not be valid for the time-scale of the simulation. The system itself consisted of a square of 500×500 lattice sites and we applied periodic boundary conditions. To exclude any affects of the boundary conditions we only calculated the VACF up to five hundred time steps. The VACF was obtained by averaging results obtained from 25 independent runs, each of which was equilibrated until $\langle u_\alpha^2 \rangle$ was independent of time. Using these parameters $D(t)$ exceeds its Boltzmann value by an order of magnitude after about thirty time steps. We illustrate this in Fig. 1 by plotting $D(t)$ at relatively short times. The data for $D(t)$ was obtained by integrating the VACF.

In Fig. 2 we have plotted values of $t\phi(t)/\sqrt{\phi(0)}$, obtained from the simulation, against t . Also shown in Fig. 2 is the theoretically predicted super long time form of

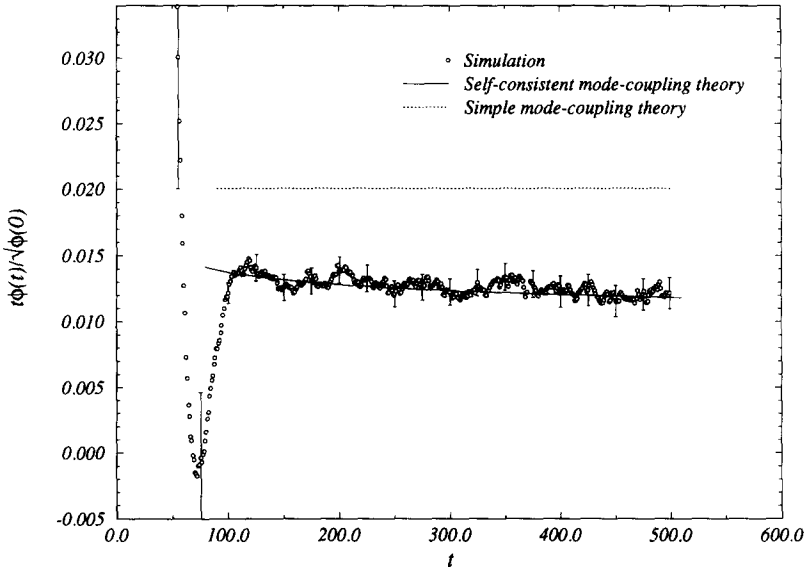


Fig. 2. The velocity autocorrelation function for the two-dimensional fluid, $\phi(t)$, multiplied by the time t , and plotted against time. The solid line is the prediction of self-consistent mode coupling theory and the broken line the prediction of the simple mode coupling theory.

this function (Eq. (5)). Over a time range starting at about 100 time steps up until the maximum time we have studied (500 time steps) our simulation data is consistent, within the errors, with the theoretically predicted asymptotic decay. This is in contrast to the simple mode coupling prediction, given in Eq. (3) and also plotted in Fig. 2, which at no point describes our data. The fact that the theoretically predicted super long time form of the VACF is consistent with our data means that the *coefficient* characterizing the long time decay must be consistent with its theoretical super long time value. Looking for this quantitative agreement is a test which can be applied more rigorously than looking specifically at the form of the deviation from $1/t$ behaviour (which according to theory should only be about 15% over the time range we have studied) and is our main quantitative evidence for super long time diffusion.

We can however attempt to quantify the deviations from $1/t$ decay by analysing our results with a little more freedom. Assuming that $t\phi(t) = \alpha(\ln(t))^\beta$ we can determine the constants α and β by plotting $\ln(t\phi(t)/\sqrt{\phi(0)})$ against $\ln(\ln(t))$. This plot is shown in Fig. 3. The first thing to note is that, although the statistics are not really good enough to conclude that the plot is linear (not to mention the fact that it is important to span a large distance in $\ln(\ln(t))$), it has a statistically significant negative slope, indicating that the decay of the VACF is faster than $1/t$. Taking the largest time range which it appears reasonable to use ($t = 110$ to $t = 500$, the range shown in Fig. 3), a linear fit to the data gives values $\alpha = 0.028 \pm 0.002$ and $\beta = -0.465 \pm 0.025$ compared with theoretical values of 0.029 and -0.5 , respectively. Because there is an uncertainty about just when the asymptotic decay sets in we have also examined the effect of varying the time at which we start to fit the data. In Table 1 we list the values of α and β

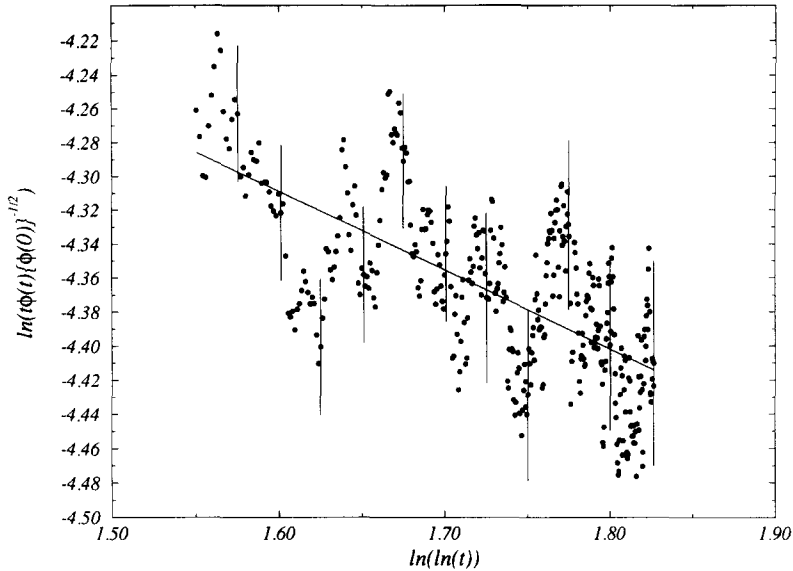


Fig. 3. Log-log plot of $t\phi(t)$ against $\ln(t)$. The solid line is a linear least squares fit to the data in the time range $t = 110 \rightarrow t = 500$.

Table 1

The parameters obtained from a linear least squares fit to the data shown in Fig. 3 for various time ranges

Time range	β	α
110 \rightarrow 500	-0.465 ± 0.025	0.028 ± 0.002
150 \rightarrow 500	-0.46 ± 0.03	0.030 ± 0.002
200 \rightarrow 500	-0.60 ± 0.05	0.040 ± 0.05
250 \rightarrow 500	-0.5 ± 0.1	0.033 ± 0.05
300 \rightarrow 500	-0.5 ± 0.2	0.03 ± 0.01

The exponent, β , and coefficient, α , are both defined in the text.

we obtained by fitting from times 110, 150, 200, 250 and 300, onwards. As Table 1 shows, there is no systematic drift in the values we obtain and the parameters are in each case, within the errors, consistent with the theoretical values. The errors do of course increase with decreasing time range because of the reduced amount of data. We did not attempt to fit the data starting at times significantly greater the 300 because at this point the errors become comparable to the quantities we are calculating, rendering the results meaningless.

4. Conclusions

By studying the dispersion of a passive tracer in a two-dimensional fluid, represented by a Lattice Boltzmann equation with induced fluctuations, we were able to satisfy the conditions necessary to see the theoretically predicted super long time decay of

the velocity autocorrelation function. Firstly, by giving the tracer a small Boltzmann diffusion coefficient the non-Boltzmann contribution, arising from the fluctuations, was made to exceed the Boltzmann value after a few time steps. Secondly, by using a Boltzmann viscosity less than the Boltzmann diffusion coefficient we were apparently able to eliminate the effects of the time dependent viscosity.

Analysing our results for the VACF we firstly concluded that our data was in quantitative agreement with the results of self-consistent mode coupling theory. Secondly, if we assumed the functional form of the decay predicted by theory and allowed both the coefficient of the logarithmic term and a pre-factor to be free parameters, then our data was best described (to within about 10%) by the theoretically expected exponent of the logarithm (an inverse square root), and *also* with the theoretically predicted pre-factor.

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