PHYSICAL REVIEW A VOLUME 27, NUMBER 6 JUNE 1983

Long-time tails and diffusion

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(Received 4 November 1982)

The evidence for long-time tails as a part of the velocity autocorrelation function for a classical particle is examined. Computer simulations, theoretical treatments, and light-scattering experiments are discussed. It is argued that numerical error propagation in the computer simulations may give rise to long-time tails as an artifact which is of hydrodynamic character. The theoretical treatments of physicists, whether avowedly phenomenological or putatively rigorous, all rely on a hydrodynamic mechanism. The usual theoretical treatments by mathematicians, based on the differential topology of "dynamical systems," appear not to be in agreement with long-time tails. Recent experiments with polystyrene spheres, observed by light scattering, claim to see the effect of long-time tails. These experiments may be interpreted as observations of the Stokes-Boussinesq effect expected for "macroscopic" spheres, but not justified for truly microscopic, molecular-sized, particles. While long-time tails may yet be rigorously established by computer simulation, theory, and experiment, it is argued in this paper that this has not happened yet. Moreover, a firm basis for serious doubt is raised.

I. INTRODUCTION

The Langevin equation[1,2] provides an elegant and successful explanation of Brownian motion.[3] The theory is quantitative and enables one to compute the diffusion constant for a macromolecule with a diameter of 10 Å. There are two ingredients for this computation which have become prototypic of the connection between correlation functions and transport coefficients generally.[4–6] The first ingredient is a consequence of the Langevin theory and gives an exponential decay for the velocity autocorrelation function of a sphere[1,2]:

$$\langle v(t)v(0)\rangle = \frac{k_B T}{M} e^{-t/\tau} \tag{1}$$

in which $v(0)$ is the initial velocity, $v(t)$ is the velocity at time t, k_B is Boltzmann's constant, T is the temperature of the fluid in which the Brownian sphere is immersed, M is its mass, and τ is the relaxation time given by Stokes' law[4]:

$$\tau = \frac{M}{6\pi\eta R} \tag{2}$$

in which η is the viscosity of the fluid and R is the radius of the sphere. The second ingredient is the Green-Kubo formula[4–6] connecting the diffusion constant D with the velocity autocorrelation function:

$$D = \int_0^\infty dt \langle v(t)v(0)\rangle = \frac{k_B T}{6\pi\eta R} . \tag{3}$$

This expression for D was already obtained by Einstein,[3] but the connection between a correlation function and a dissipation coefficient, arrived at independently by Green and Kubo, is of much more general validity.

Early theoretical work based on the Boltzmann equation, or on the Boltzmann-Enskog equation, provided justification for the exponential decay law[6,7] for the velocity autocorrelation function in (1). This was the basis for the belief in separated time scales during dynamical evolution in many-particle systems, and became the impetus behind the Bogoliubov-Born-Green-Kirkwood Yvon (BBGKY) hierarchy approach to rigorous kinetic theory.[8]

It was a great surprise at the end of the 1960's when Alder and Wainwright[9] reported that computer simulations of hard spheres and hard disks gave rise to a velocity autocorrelation function possessing an asymptotic power law, a "long-time tail," instead of an exponential decay:

$$\langle v(t)v(0)\rangle \sim t^{-d/2} \quad \text{for} \quad t \to \infty \tag{4}$$

in which d is the dimensionality of the system simulated. Nevertheless, within a very short time, several theoretical explanations appeared which explained the computer results and corroborated each other.

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A nonlinear approach, now called "mode-coupling" theory, was presented by Bedeaux and Mazur.[10] The method was independently used by Kawasaki[11] somewhat earlier. The mode-coupling approach was reviewed by Pomeau and Résibois[12] as early as 1975. A linear, phenomenological approach, strongly dependent on hydrodynamics, and already evident in Kawasaki's work, was presented by several authors.[13] One of these authors, Hauge,[14] also invoked the Stokes-Boussinesq theory[15–17] which goes back to 1903. Each of these approaches is avowedly phenomenological in one way or another. Dorfman and Cohen[18] provided the first putatively rigorous, kinetic-theory approach in a pair of papers, which appeared in 1972 and 1975. Their results agreed with the other approaches, and with the computer simulations. Some physicists viewed the problem as essentially solved at this point, and subsequent textbooks treat it as such. Wood and Erpenbeck[19] carefully reproduced and extended the computer simulations. However, light-scattering experiments were only reported much more recently.[20] These experiments support the long-time–tail viewpoint, but their interpretation requires careful discussion and will be so treated later in this paper.

Parallel but independent of the development described above, progress continued in an entirely different arena. The very mathematical approach[21] to "dynamical systems," which depends heavily on differential topology and differential geometry, also produced surprising results. Chief among these were the results on "strange attractors" which appear in dissipative dynamical systems. Application of the methods involved to Hamiltonian dynamics is also possible[22,23] and provides the modern form of the old ergodic theory ideas.[24] In the cases of hard spheres (billiards), and the Lorentz gas, very recent results for the velocity autocorrelation function have been achieved by Bunimovich and Sinai.[25] These results very much sharpen the conditions required for long-time tails or their absence, and appear to vitiate a great deal of work by physicists on the Lorentz-gas model.[26] Moreover, criticisms of the computer simulations based on numerical error propagation have been answered by some by invocation of the so-called "β-shadow" theorem. This theorem is a by-product of the same mathematical approach just described, and its applicability to the computer simulations for hard spheres is open to serious question.[27]

Because the issue of long-time tails is currently of central importance in nonequilibrium statistical mechanics, and because there are several grounds for doubt, it is the purpose of this paper to establish these doubts in some detail. It is hoped that this will stimulate others to answer these doubts, and thereby sharpen or definitively settle the issue. It is not our purpose to claim that long-time tails in diffusion processes are an artifact of numerical error propagation. This possibility needs to be explored, and cannot be ruled out on the basis of present evidence from computer simulation, theory, or experiment.

The paper is organized as follows:

In Sec. II the computer simulations for the velocity autocorrelation function are described. How numerical error propagation could affect the results is discussed. In particular, it is argued that velocity *cross*correlations rather than *auto*correlations are being simulated, and that these correlations lead to hydrodynamic correlations. The β-shadow theorem for Anosov systems is discussed, as well as the question of whether or not the billiard systems being simulated are in fact Anosov systems.

In Sec. III the mode-coupling theory and results are summarized. It is observed that the results are obtained from hydrodynamic velocity field correlations and not from particle autocorrelations. The connection between this observation and the discussion of Sec. II is elucidated.

In Sec. IV both the phenomenological linearized hydrodynamic theory and the rigorous kinetic theory are discussed. It is shown that both obtain the long-time tail from hydrodynamic correlations identical with those invoked in Secs. II and III.

In Sec. V exact results are presented for coupled harmonic oscillators. These are the only exact results available, and their marginal significance for the general problem is discussed.

In Sec. VI the Stokes-Boussinesq explanation of long-time tails is summarized. The usual treatment is criticized, modified, and discussed in detail. Recent experiments involving light scattering from polystyrene spheres are discussed and interpreted in terms of the Stokes-Boussinesq theory. It is argued that this does not imply identical results for truly microscopic molecules, contrary to widespread belief.

In Sec. VII the results of studies on the Lorentz-gas model are summarized. The idea of a "finite horizon," due to Sinai, is discussed. A possible incompatibility of the physicist's and the mathematician's approach is adumbrated.

In Sec. VIII an elementary statement of the results of the modern theory of dynamical systems is given. Several problems related to the issue of long-time tails are mentioned in addition to those issues already raised in preceding sections. It is argued that this branch of mathematics needs to be much more closely associated with the efforts in kinetic theory.

In Sec. IX concluding remarks are presented.

II. COMPUTER SIMULATIONS

The computer simulations of Alder and Wainwright[9] and of Wood and Erpenbeck[19] are capable of simulating the behavior of several hundred to perhaps a few thousand hard disks or hard spheres. To make such small systems have properties approximating those of systems of macroscopic size, periodic boundary conditions are used.[19] The simulations run typically to $20t_0$ or $30t_0$, where t_0 is the mean free time. By $10t_0$ or $12t_0$, the exponential portion of the velocity autocorrelation decay is replaced by the long-time—tail power-law decay.

Ideally, the computed trajectory would correspond with the real Newtonian trajectory at least for the entire time course of the simulation. We would want to be able to stop the simulation after, for example, $30t_0$, reverse all velocities, and then backevolve the trajectory to $t=0$. Numerical accuracy would be achieved if the backevolved trajectory ended up within a sufficiently small ϵ neighborhood of the initial conditions for the original forward evolution. In the computer simulations so far, this objective is not achievable. Erpenbeck[28] has stated the following: "The ability of molecular-dynamics calculations to evaluate the velocity autocorrelation function to relatively long times ($\sim 30t_0$, where t_0 is the mean free time) is, of course, subject to question, especially for values of the time greater than that for which an individual trajectory loses essentially all its accuracy [$\sim 12t_0$ for 48-bit (binary digit) (single-) precision arithmetic for moderately dense hard disks]."

This problem of numerical error propagation in computer simulations has been appreciated for some time. Many practitioners[28,29] invoke the so-called β-shadow theorem[30] to justify their empirical observation that time averages obtained in simulations are essentially independent of numerical methods used and machines used. This theorem, independently proved by Anosov[31] and Bowen,[32] arises in the mathematical theory of dynamical systems. It is proved for Axiom-A systems,[32] which includes the Anosov systems.[33]

The Anosov-Bowen theorem may be expressed as follows. Let T be a mapping on a compact manifold M. T is *assumed* to be an Axiom-A diffeomorphism. Let x denote a point in M. A sequence of points $\{x_i\}$, where $i=1,2,\ldots,$ in M is an α *pseudo orbit* if

$$d(Tx_i,x_{i+1})<\alpha \quad \text{for all} \quad i\ , \tag{5}$$

in which d is a distance measure on M. A point x "β shadows" the sequence $\{x_i\}$ if

$$d(T^i x,x_i)\leq\beta \quad \text{for all} \quad i=1,2,\ldots \tag{6}$$

in which T^i is the ith iterate of T. The Anosov-Bowen theorem can now be stated:

For every $\beta\to 0$ there is an $\alpha>0$ so that every α pseudo orbit $\{x_i\}$, $i=1,2,\ldots,$ in M is β shadowed by a point x in M.

Application of this theorem to computer simulations of hard-sphere (disk) systems is achieved by identifying M with phase space and identifying T as the Hamiltonian evolution in phase space for a fixed discrete time interval. All we have to do is choose the time interval small enough so that the numerical accuracy of the computer will guarantee the condition (5) for an α pseudo orbit. Thus, the numerical trajectory is an α pseudo orbit, but it is β shadowed by a true Newtonian (Hamiltonian) orbit.

There is a catch to the application of the Anosov-Bowen theorem. We have not verified that the hard-sphere system is an Anosov system (or Axiom-A). Sinai has proved that the system is a K system,[34] and he has also shown that hard-sphere systems are *not* Anosov.[27] So we really do not know whether the Anosov-Bowen theorem is applicable. Moreover, the failure of the hard-sphere system to be Anosov is a result of a technicality,[27] which in the present context may be crucial. Anosov systems are said to be exponentially unstable,[21,32] i.e., the T flow on M is "hyperbolic." For hard spheres, the exponential instability is not "uniform," i.e., there is no lower bound on the exponential rate. On the one hand, if the hard-sphere system were Anosov, then exponential instability in M would suggest (nothing has been *proved* here) that the velocity autocorrelation of a single sphere should decay exponentially. On the other hand, the failure of the hard-sphere system to be Anosov results from the lack of a lower bound on the exponential rate in M, and this situation is what can give rise to long-time tails in other model systems containing only *one* moving particle.[35] Ruell[21] has stated that there is evidence that correlations decay exponentially for Axiom-A flows; but at the time of his writing this characterization was unproved, and an outstanding open question. We shall return to this issue in Sec. VII during the discussion of the Lorentz-gas model.

It is important to distinguish between trajectories in phase space and the trajectories of individual particles in ordinary three-dimensional (two-dimensional) space. Clearly, if the phase-space trajectory moves away from the true, Newtonian trajectory, then the projected motion of individual particles will also become numerically erroneous. A detailed account of the connection between these two perspectives is not to be found in the extant literature

on Axiom-A diffeomorphisms. Consider a tagged sphere (disk) and its 3D (2D) trajectory. Initially, the computer simulation keeps the numerical trajectory coincident with the Newtonian trajectory. Around $10t_0$ these two trajectories become different. It is possible that the numerical trajectory subsequently becomes very similar with the Newtonian trajectory of another particle (remember—the Anosov-Bowen theorem is for phase space, and we are now in 3D space). As the numerical trajectory continues to evolve, it changes from one individual Newtonian trajectory to another and another and so on. Consequently, the computer-determined velocity correlation function is not really an *auto*correlation function, but is perhaps quite close to being a *mutual*-correlation (or *cross*-correlation) function for many, different, individual particles and their associated velocities. Let the tagged particle velocity be denoted by $v_1(t)$. We want $\langle v_1(t)v_1(0)\rangle$ for all t, but we are really getting something more like

$$\begin{aligned}
&\langle v_1(t)v_1(0)\rangle \quad \text{for} \quad t\in(0,t_1)\ ,\\
&\langle v_2(t)v_1(0)\rangle \quad \text{for} \quad t\in(t_2,t_3)\ ,\\
&\langle v_3(t)v_1(0)\rangle \quad \text{for} \quad t\in(t_4,t_5)\ ,\\
&\vdots\\
&\langle v_n(t)v_1(0)\rangle \quad \text{for} \quad t\in(t_{2n-2},t_{2n-1})\ ,
\end{aligned} \tag{7}$$

where the numerical trajectory agrees with the Newtonian trajectory for particle n during the interval (t_{2n-2},t_{2n-1}). During the gaps (t_{2n-1},t_{2n}), the numerical trajectory may not be close enough to any particular particle trajectory.

Mutual correlations do occur when one deals with fluids.[6,7] The hydrodynamic velocity *field* is the averaged sum of the velocities of all those particles in a mesoscopic volume centered at the field point $\vec{r}$:

$$\rho(\vec{r},t)\vec{u}(\vec{r},t)=\sum_i \vec{v}_i\delta(\vec{r}-\vec{r}_i(t))\ . \tag{8}$$

The velocity *field auto*correlation will be made up of both labeled particle velocity autocorrelations and mutual correlations. The latter will dominate by virtue of being of order N^2 whereas the former are of order N, where N is the number of particles in a mesoscopic fluid element (N is typically 10^{12}). A straightforward calculation based on fluctuating hydrodynamics[36] yields

$$\langle u_\alpha(\vec{r},t)u_\beta(\vec{r}',t')\rangle=\tfrac{2}{3}k_BT\frac{1}{\rho}(4\pi\nu\,|\,t-t'\,|\,)^{-3/2}\delta_{\alpha\beta} \tag{9}$$

in the limit

$$\frac{\nu^{1/2}\,|\,t-t'\,|^{1/2}}{|\,\vec{r}-\vec{r}'\,|}\to\infty\ , \tag{10}$$

where ν is the kinematic viscosity, ρ is the mass density, and α and β are indices for cartesian components of the velocity field. The limit (10) means that the velocity field fluctuations are in a local region for *long* times. This "hydrodynamic" correlation, which is *not* a tagged particle velocity autocorrelation, does yield a long-time tail. Could it be that numerical error propagation changes autocorrelations into mutual correlations characteristic of hydrodynamic correlations, which possess long-time tails? If this is so, then the long-time tails observed in the computer simulations are not necessarily those of a tagged particle, but are instead those associated with the hydrodynamiclike numerical noise. This perspective leaves entirely open the question of whether the tagged particle velocity autocorrelation function *also* possesses a long-time tail anyway.

III. MODE COUPLING

The mode-coupling theory[10–12] of the long-time tails begins with the hydrodynamic equations for a binary mixture. The species whose diffusion constant is of interest to us is a dilute solute species in some solvent. The solute is represented by a concentration $c(\vec{r},t)$ which satisfies the nonlinear equation[10]

$$\frac{\partial}{\partial t}c(\vec{r},t)=D_0\nabla^2c(\vec{r},t)-\vec{u}(\vec{r},t)\cdot\vec{\nabla}c(\vec{r},t) \tag{11}$$

in which D_0 is the "bare" diffusion constant, and $\vec{u}(\vec{r},t)$ is the velocity field of the mixture. The term containing $\vec{u}(\vec{r},t)$ is the nonlinear, mode-coupling term. Since the solute is dilute, $\vec{u}(\vec{r},t)$ can be thought of as essentially the velocity field of the solvent alone. It satisfies the linearized, fluctuating Navier-Stokes equation[36]

$$\begin{aligned}
\frac{\partial}{\partial t}u_\alpha(\vec{r},t)=&-\frac{1}{\rho}\frac{\partial}{\partial x_\alpha}p+\nu\nabla^2u_\alpha(\vec{r},t)\\
&+\frac{1}{\rho}\frac{\partial}{\partial x_\beta}\tilde{S}_{\alpha\beta}(\vec{r},t)
\end{aligned} \tag{12}$$

in which p is the pressure and $\tilde{S}_{\alpha\beta}$ is the fluctuating stress tensor.[36] The summation over β is implicit. The mode-coupling strategy is to solve (12) for $\vec{u}(\vec{r},t)$. This quantity fluctuates because of $\tilde{S}_{\alpha\beta}$. Therefore, in (11), the nonlinear term becomes a multiplicative stochastic[36] term and $c(\vec{r},t)$ satisfies a multiplicative stochastic differential equation. Equation (11) is averaged with respect to the fluctuating velocity field $\vec{u}(\vec{r},t)$ by means of cumulants.[36] In the limit $|\,\vec{k}\,|\to0$, where $\vec{k}$ is the spatial Fourier-transform wave vector, the "renormalized" diffusion equation is obtained:

$$\frac{\partial}{\partial t}\langle c(\vec{r},t)\rangle = D_0\nabla^2\langle c(\vec{r},t)\rangle + \frac{2}{3}\frac{k_BT}{\rho}\int_0^t ds[4\pi(D_0+\nu)\,|\,t-s\,|\,]^{-3/2}\nabla^2 c(\vec{r},t)\ . \tag{13}$$

In this analysis, it is not the velocity autocorrelation function for a *tagged* particle which has been used to calculate the renormalized diffusion coefficient,

$$D_R = \frac{2}{3}\frac{k_BT}{\rho}\int^t ds[4\pi(D_0+\nu)\,|\,t-s\,|\,]^{-3/2}\ , \tag{14}$$

but the hydrodynamic velocity *field autocorrelation* function. The averaging procedure which converts (11) into (13) depends, in the $|\vec{k}|\to 0$ limit, on the second cumulant, which is determined precisely by the second moment of $\vec{u}(\vec{r},t)$, i.e., the velocity *field autocorrelation* function. This is why the integrand in (13) and (14) is so similar to Eq. (9). The difference is that the factor ν in (9) becomes $D_0+\nu$ in (13) and (14). This is a result of the mixing of the two terms on the right-hand side of (11) during the averaging procedure. At room temperature, with water as the solvent, ν is at least 10^3 times bigger than D_0 for all molecules of radius bigger than 3 Å. Consequently, this difference is effectively unobservable.

The long-time tail has arisen from hydrodynamic fluctuations. The agreement with the computer simulations could be interpreted as further support for the notion that numerical noise is of hydrodynamic character, rather than as support for the view that a tagged particle's velocity autocorrelation function has a long-time tail. The agreement is not fortuitous; rather, the explanation fits the cause of the observed "artifact." It may be so that the tagged particle's velocity autocorrelation function has a long-time tail, but this mode-coupling agreement does not establish this proposition.

The fact that hydrodynamic fluctuations give rise to long-time tails has surfaced in both this section and in Sec. II. In Sec. IV more will be said about how it occurs and, in particular, it will be shown to be a consequence of the diffusive character of the Navier-Stokes equations.

The mode-coupling theory may be criticized on another basis. The structure of Eqs. (11) and (12) which leads to the interpretation of (11) as a multiplicative stochastic differential equation may not be correct. Macroscopic equations, such as the hydrodynamic equations, are expressed in terms of macroscopic quantities, such as $c(\vec{r},t)$ and $\vec{u}(\vec{r},t)$, which are obtained from microscopic particle dynamics by averaging procedures.[7,8] One then obtains equations which are perhaps nonlinear in the averaged, macroscopic quantities. The equations for the fluctuations of these macroscopic quantities satisfy equations which are obtained by linearization of the averaged equations and which contain additive stochastic inhomogeneous terms.[36] At least this is what happens if any sort of master equation approach is used.[36–39] Equation (12) is the kind of equation which does appear in such treatments. Equation (11) with a stochastic interpretation for $\vec{u}(\vec{r},t)$ is not. There is a theory for the fluctuations of a binary mixture,[40] and it just is not of the form of (11) and (12) with the interpretation given above. I think the attractiveness of the mode-coupling approach to long-time tails stems from its apparent agreement with the computer simulation results. As noted above, this may be illusory.

IV. KINETIC THEORY

An exact calculation of the velocity autocorrelation function for a tagged particle requires solving the N-body problem exactly. This is, in general, not possible. Consequently, a rigorous approach based on kinetic theory must necessarily involve approximations which, it is hoped, are well controlled. The work of Ernst *et al.*,[13] was avowedly phenomenological, but it provided the outline for a rigorous approach in which the phenomenological steps could, perhaps, be replaced by rigorous kinetic-theory arguments. The tour de force attack of Dorfman and Cohen,[18] using kinetic theory, went further in this direction than one previously imagined possible. Some textbook authors[41] have even treated their work as the definitive conclusion to the whole development. In this section of this paper, the mechanism behind the work of Ernst *et al.*, and the work of Dorfman and Cohen will be discussed. Once again we will see that it is hydrodynamics which gives rise to the long-time tail. Moreover, the theory of Dorfman and Cohen will be seen to involve several approximations or truncations, which cannot be said to be well controlled in any rigorous sense.

The velocity autocorrelation function for a tagged particle in an N-particle system can be expressed as

$$\begin{aligned}\rho_D(t) &= \tfrac{1}{3}m\beta\langle\vec{v}_1(t)\cdot\vec{v}_1(0)\rangle \\ &= \tfrac{1}{3}m\beta\langle\vec{v}_1(0)\cdot\vec{v}_1(-t)\rangle\end{aligned} \tag{15}$$

in which m is the mass of the tagged particle, $\beta = 1/k_BT$, and time translation invariance has been used. The average can be written

$$\langle \vec{v}_1(0)\cdot\vec{v}_1(-t)\rangle = \langle \vec{v}_1\cdot e^{-tL_N}\vec{v}_1\rangle = \frac{1}{Z}\int d^N x\, \vec{v}_1\cdot e^{-tL_N}e^{-\beta H_N}\vec{v}_1 \tag{16}$$

in which $Z^{-1}e^{-\beta H_N}$ is the canonical distribution and L_N is the Liouville operator

$$L_N = \sum_{i=1}^{N}\frac{\vec{p}_i}{m}\cdot\frac{\partial}{\partial\vec{r}_i} - \sum_{i<j}\left[\frac{\partial\phi(r_{ij})}{\partial\vec{r}_i}\cdot\frac{\partial}{\partial\vec{p}_i} - \frac{\partial\phi(r_{ij})}{\partial\vec{r}_j}\cdot\frac{\partial}{\partial\vec{p}_j}\right] \tag{17}$$

while H_N is the Hamiltonian

$$H_N = \sum_{i=1}^{N}\frac{p_i^2}{2m} + \sum_{i<j}\phi(r_{ij}) \tag{18}$$

in which $r_{ij} = |\vec{r}_i - \vec{r}_j|$. The integral in (16) is over all coordinate and momenta and is symbolized by $d^N x = d^6x_1 d^6x_2\cdots d^6x_N$, where $d^6x_i = d^3r_i d^3p_i$.

In the phenomenological treatment, the exact time evolution implicit in (16) is replaced by an approximation which is essentially the hydrodynamic relaxation of local equilibrium. The result is an expression with the form[42]

$$\rho_D(t) \simeq \tfrac{1}{3}m\beta\int d^3v_0\int d^3r\,\vec{v}_0\cdot P(\vec{r},t)\vec{U}(\vec{r},t)\phi_0(\vec{v}_0) \tag{19}$$

in which $\phi_0(\vec{v}_0)$ is the normalized Maxwell distribution and $P(\vec{r},t)$ and $\vec{U}(\vec{r},t)$ are the probability of finding the tagged particle and the local velocity at $\vec{r}$ at time t, respectively. The relaxation of $P(\vec{r},t)$ is governed by a diffusion equation

$$\frac{\partial}{\partial t}P(\vec{r},t) = D\nabla^2 P(\vec{r},t) \tag{20}$$

whereas the relaxation of $\vec{U}(\vec{r},t)$ is governed by the diffusionlike Navier-Stokes equation

$$\frac{\partial}{\partial t}\vec{U}(\vec{r},t) = \nu\nabla^2\vec{U}(\vec{r},t) + \tfrac{1}{3}\nu\vec{\nabla}[\vec{\nabla}\cdot\vec{U}(\vec{r},t)] \tag{21}$$

in which ν is the kinematic viscosity. Equation (21) neglects compressibility contributions, such as bulk viscosity and sound propagation, which are not of direct concern in the present context. The initial conditions for (20) and (21) are

$$P(\vec{r},0) = W(\vec{r}-\vec{r}_0)$$

and (22)

$$\vec{U}(\vec{r},t) = \frac{m}{\rho}\vec{v}_0 W(\vec{r}-\vec{r}_0)$$

in which $W(\vec{r}-\vec{r}_0)$ is a distribution of initial positions for the tagged particle, $\vec{v}_0$ is the initial velocity, and $m^{-1}\rho$ is the mean particle density. Introduction of Fourier transforms

$$P(\vec{k},t) = \int d^3r\, P(\vec{r},t)e^{i\vec{k}\cdot\vec{r}}$$

and (23)

$$\vec{U}(\vec{k},t) = \int d^3r\,\vec{U}(\vec{r},t)e^{i\vec{k}\cdot\vec{r}}$$

converts (19) into

$$\rho_D(t) \simeq \frac{2}{3}\frac{m}{\rho}\int d^3k\frac{1}{(2\pi)^3}W(\vec{k})W(-\vec{k})e^{-(D+\nu)k^2t} \tag{24}$$

in which $W(\vec{k})$ is the Fourier transform of $W(\vec{r}-\vec{r}_0)$, the solutions to (20) and (21) have been used, and the Maxwell velocity average has already been executed as a result of the additional factor of $\vec{v}_0$ in (22). This result is expected to be valid only for long times and for small initial spatial inhomogeneities, which are introduced by $W(\vec{r}-\vec{r}_0)$. Consequently, the $W(\vec{k})W(-\vec{k})$ factor is replaceable by 1, and the resulting integral yields

$$\rho_D(t) \simeq \frac{2}{3}\frac{m}{\rho}[4\pi(D+\nu)t]^{-3/2}. \tag{25}$$

The similarity with (9) is, of course, striking. Comparison with (14) provides even stronger support for the conclusion that the argument is, in essence, correct.

In view of the discussions in Secs. II and III, we could argue instead that we are getting long-time tails once again because we have used a hydrodynamic argument which is guaranteed to reproduce the effects of numerical noise because of their hydrodynamiclike character. In fact, we see that the long-time tails come from the diffusive nature of the hydrodynamic equations. This can also be exhibited for the case of the hydrodynamic velocity field correlation (9) which follows from the integral

$$\langle u_\alpha(\vec{r},t)u_\beta(\vec{r}',t')\rangle = \frac{k_B T}{\rho}\left[-\nabla^2\delta_{\alpha\beta} + \frac{\partial}{\partial x_\alpha}\frac{\partial}{\partial x_\beta}\right]\frac{1}{(2\pi)^3}\int d^3k\frac{e^{-i\vec{k}\cdot(\vec{r}-\vec{r}')}}{k^2}e^{-\nu k^2|t-t'|}. \tag{26}$$

In the limit $|\vec{r}-\vec{r}'|\to 0$, this becomes just

$$\langle u_\alpha(\vec{r},t)u_\beta(\vec{r},t')\rangle = \frac{2}{3}\frac{k_BT}{\rho}\frac{4\pi}{(2\pi)^3}\int_0^\infty dk\, k^2 e^{-\nu k^2|t-t'|}$$
$$= \frac{2}{3}\frac{k_BT}{\rho}(4\pi\nu|t-t'|)^{-3/2} . \tag{27}$$

The Gaussian character of this integral and the integral in (24) is a result of diffusive dynamics. Thus, the hydrodynamic character of numerical noise in the computer simulations is really of a diffusive nature, *provided* that the hypothesis that it is the noise which produces the long-time tail is correct.

It is interesting to note that in Sec. II it was observed that hard-sphere systems failed to be Anosov systems because the exponential instability was not uniform, i.e., there is no lower bound on the exponential rate. In integrals (24) and (27) the long-time tail results from an identical effect. The exponential decay of the integrand does not have a lower bound either, since $k\to 0$ is allowed. Therefore, correlations of diffusive origin are incompatible with Anosov behavior. In Sec. VI particularly, it will be argued that the diffusive modeling of the true underlying microscopic dynamics is especially poor for short times, as is well known, and that this causes the long-time behavior of the correlation functions. One could also argue that the diffusive behavior is consistent with the nonAnosov character of hard-sphere systems. The negation of this assertion will be explored in Sec. VII.

The kinetic-theory approach to the calculation of (16) does not involve wholesale substitutions as in the phenomenological approach just described. Instead, it purports to be a rigorous calculation modulo certain hopefully innocuous simplifications.

The procedure used is that of a hierarchy which is truncated. Moreover, ultimately the analysis is specialized to hard-sphere interactions, which are of a singular nature when viewed from the potential form of (17). It is possible to introduce a modified, but exact, form of the Liouville operator L_N which describes the dynamics for hard spheres. This is given by

$$\bar{L}_N = \sum_{i=j}^{N}\frac{\vec{p}_i}{m}\cdot\frac{\partial}{\partial\vec{r}_i} - \sum_{i<j}\bar{T}(i,j) \tag{28}$$

in which $\bar{T}(i,j)$ is the binary-collision operator.[18,43] For hard spheres, the velocity autocorrelation in (16) is modified by replacing L_N with $\bar{L}_N$. Equation (15) is rewritten as

$$\rho_D(t) = \int d^3v_1\,\vec{v}_1\cdot\vec{\Phi}_D(\vec{v}_1,t) , \tag{29}$$

where $\vec{\Phi}_D(\vec{v}_1,t)$ is defined by

$$\vec{\Phi}_D(\vec{v}_1,t) = \tfrac{1}{3}m\beta\frac{V}{Z}m^3\int d^6x_2\cdots d^6x_N e^{-t\bar{L}_N}e^{-\beta H_N}\vec{v}_1 \tag{30}$$

in which the factor of V came from the d^3r_1 integration, since only relative coordinates appear in the integrand. The hierarchy is derived by looking at $(\partial/\partial t)\vec{\Phi}_D$:

$$\frac{\partial}{\partial t}\vec{\Phi}_D(\vec{v}_1,t) = \frac{\rho}{m}\int d^3r_2 d^3v_2\,\bar{T}(1,2)\vec{\Phi}_D(x_1,x_2,t) , \tag{31}$$

where

$$\vec{\Phi}_D(x_1,x_2,t) = \tfrac{1}{3}m\beta\frac{V^2}{Z}m^6 \times\int d^6x_3\cdots d^6x_n e^{-t\bar{L}_N}e^{-\beta H_N}\vec{v}_1 . \tag{32}$$

Next, $(\partial/\partial t)\vec{\Phi}_D(x_1,x_2,t)$ is given by

$$\frac{\partial}{\partial t}\vec{\Phi}_D(x_1,x_2,t)+\bar{L}_2(1,2)\vec{\Phi}_D(x_1,x_2,t) = \frac{\rho}{m}\int d^3r_3 d^3v_3[\bar{T}(1,3)+\bar{T}(2,3)] \times\vec{\Phi}_D(x_1,x_2,x_3,t) , \tag{33}$$

where

$$\vec{\Phi}_D(x_1,x_2,x_3,t) = \tfrac{1}{3}m\beta\frac{V^3}{Z}m^9\int d^6x_4\cdots d^6x_n e^{-t\bar{L}_N}e^{-\beta H_N}\vec{v}_1 . \tag{34}$$

This process is continued indefinitely until an infinite hierarchy is produced. In each expression, we have in mind, implicitly, the thermodynamic limit: $N\to\infty$, $V\to\infty$, $N/V=\rho/m$. The direct computation of (29) has been converted into an infinite set of coupled, first-order differential equations.

The next step of the procedure is to introduce cluster functions. These are defined by

$$\vec{\Phi}_D(x_1,x_2,t)=\vec{\Phi}_D(\vec{v}_1,t)\phi_0(\vec{v}_2)+\vec{\chi}_D(x_1,x_2,t)\ ,$$
$$\vec{\Phi}_D(x_1,x_2,x_3,t)=\vec{\Phi}_D(\vec{v}_2,t)\phi_0(\vec{v}_2)\phi_0(\vec{v}_3)+\vec{\chi}_D(x_1,x_2,t)\phi_0(\vec{v}_3)+\vec{\chi}_D(x_2,x_3,t)\phi_0(\vec{v}_2)+\vec{\chi}_D(x_1,x_2,x_3,t)\ , \tag{35}$$
$$\ldots .$$

The hierarchy now takes the form

$$\frac{\partial}{\partial t}\vec{\Phi}_D(\vec{v}_2,t)=\frac{\rho}{m}\int d^3r_2d^3v_2\bar{T}(1,2)[\vec{\Phi}_D(\vec{v}_2,t)\phi_0(\vec{v}_2)+\vec{\chi}_D(x_1,x_2,t)]\ , \tag{36}$$

$$\frac{\partial}{\partial t}\vec{\Phi}_D(x_1,x_2,t)+\bar{L}_2(1,2)\vec{\chi}_D(x_1,x_2,t)$$
$$=\bar{T}(1,2)\vec{\Phi}_D(\vec{v}_1,t)\phi_0(\vec{v}_2)+\frac{\rho}{m}\int d^3r_3d^3v_3\{\bar{T}(1,3)\phi_0(\vec{v}_3)\vec{\chi}_D(x_1,x_2,t)$$
$$+\bar{T}(2,3)(1+P_{23})\phi_0(\vec{v}_3)\vec{\chi}_D(x_1,x_2,t)$$
$$+[\bar{T}(1,3)+\bar{T}(2,3)]\vec{\chi}_D(x_1,x_2,x_3,t)\} \tag{37}$$

in which P_{23} is a permutation operator for variables labeled with 2 and 3. The strategy[18] now is to neglect $\vec{\chi}_D(x_1,x_2,t)$ in (36) and then solve the resulting closed equation for $\vec{\phi}_D(\vec{v}_1,t)$. This provides a first approximation for $\vec{\Phi}_D(\vec{v}_1,t)$, which is then used in (37) where $\vec{\chi}_D(x_1,x_2,x_3,t)$ is neglected. This gives a closed equation for $\vec{\chi}_D(x_1,x_2,t)$ which provides an approximation for $\vec{\chi}_D(x_1,x_2,t)$. This approximation can be put back into (36) to get a better approximation for $\vec{\Phi}_D(\vec{v}_1,t)$. One then moves up the hierarchy in a similar fashion, all the while refining the approximate solutions for the cluster functions.

The lowest-order approximation for $\vec{\Phi}_D(\vec{v}_1,t)$ obtained in this fashion is nothing other than the Lorentz-Boltzmann equation, and it yields

$$\rho_{D,1}(t)=\exp\left[-\frac{\rho}{m}t(m\beta D_{00})^{-1}\right] \tag{38}$$

in which D_{00} is the self-diffusion coefficient from the Lorentz-Boltzmann equation in the first Enskog approximation. The subscript 1 in (38) refers to the fact that this solution is for the lowest-order approximation to the hierarchy. Equation (38) exhibits the exponential decay of the velocity autocorrelation function typically found from the Boltzmann-type equations (for example, Lorentz-Boltzmann and Boltzmann-Enskog).

The next step in the solution to the hierarchy provides an approximation to $\vec{\chi}_1(x_1,x_2,t)$. This approximation is then inserted into (36) to yield a correction to the Lorentz-Boltzmann equation. The solution to this new equation is much more involved[44] than the solution to the Lorentz-Boltzmann equation itself. In fact, the solution requires additional expansions into infinite series which can only be analyzed to the first few lowest orders. This analysis, while very demanding, has been carried out in detail,[18,44] and yields long-time tails. One does not need to go to $\vec{\chi}_D(x_1,x_2,x_3,t)$ in order to get this effect.

Because the approximation scheme begins with the Lorentz-Boltzmann equation for hard spheres, the expressions obtained are expressed in terms of the eigenfunctions and eigenvalues of the Lorentz-Boltzmann equation. The problem is analyzed through spatial Fourier transforms and temporal Laplace transform. Long times and gentle spatial variations correspond with the small Laplace-transform variable ϵ and the small Fourier-transform wave vector $\vec{k}$. In the limit $\epsilon\to 0$ and $|\vec{k}|\to 0$, the eigenfunctions and eigenvalues of the Lorentz-Boltzmann equation which dominate the approximate solution to the hierarchy based upon $\vec{\chi}_D(x_1,x_2,t)$ are the so-called "hydrodynamic modes." These arise directly from the conservation of linear momentum and kinetic energy during hard-sphere collisions. They ultimately[44] give rise to an expression for $\rho_{D,2}(t)$ which involves an integral just like that in (24). Thus, once again, the long-time tail arises from hydrodynamic behavior of diffusive character exhibited in Gaussian integrals over Fourier variables.

There is no doubt that the beautiful analysis of Dorfman[18,44] and Cohen has explained the phenomenological analysis of Ernst *et al.*[13] Nevertheless, one may well ask what would happen if $\vec{\chi}_D(x_1,x_2,x_3,x_4,x_5,t)$, for example, were included

in the analysis, or even much higher-order cluster functions. Even the analysis bases on $\vec{\chi}_D(x_2,x_2,t)$ required approximations and truncations. These appear to be reasonably controlled, but the neglect of all higher-order cluster functions has an effect on the final result which cannot reasonably be intuited. Moreover, a shift away from hard spheres towards more realistic particle interactions renders even the analysis of $\vec{\chi}_D(x_1,x_2,t)$ considerably more difficult and it removes the natural emergence of the hydrodynamic modes.

V. COUPLED OSCILLATORS

In a sense, it is inappropriate to include a discussion of coupled oscillators in this paper because they describe a solid lattice rather than a gas or liquid. Nevertheless, there are two good reasons for doing so. Coupled oscillators provide virtually the only exactly computable model for the velocity autocorrelation function. This feature was exploited early by Montroll[45] and later by Ford, Kac, and Mazur.[46] In addition, the proposition that conservation of momentum causes the long-time tail in the velocity autocorrelation function can be explicitly tested with oscillators.

Consider N coupled oscillators, each of which has mass m. The Hamiltonian for this system is given by

$$H_N=\sum_{i=1}^{N}\frac{p_i^2}{2m}+\sum_{i<j}\tfrac{1}{2}\kappa_{ij}(q_i-q_j)^2$$

with (39)

$$\kappa_{ij}=\kappa_{ji}\ .$$

In this case, the Liouville operator in (17) may be written

$$L_N=\sum_{i=1}^{N}\frac{p_i}{m}\frac{\partial}{\partial q_i}-\sum_{i<j}\kappa_{ij}(q_i-q_j)\left[\frac{\partial}{\partial p_i}-\frac{\partial}{\partial p_j}\right]. \tag{40}$$

The mutual-correlation matrix analogue of the velocity autocorrelation function in (16) may be written

$$\begin{aligned}M_{ij}(t)&\equiv\langle v_i(0)v_j(-t)\rangle\\&=\langle v_i(0)e^{-tL_N}v_j\rangle\\&=\frac{1}{Z}\int d^Nx\,v_ie^{-tL_N}e^{-\beta H_N}v_j\ .\end{aligned} \tag{41}$$

Ultimately, we will want to look at a diagonal element of this matrix because it will correspond with a velocity autocorrelation function. Now, observe that

$$\begin{aligned}\frac{d}{dt}M_{ij}(t)&=-\langle v_iL_Ne^{-tL_N}v_j\rangle\\&=-\sum_{i<l}\frac{1}{m}\kappa_{il}\langle(q_i-q_l)e^{-tL_N}v_j\rangle\\&\quad+\sum_{l<i}\frac{1}{m}\kappa_{li}\langle(q_l-q_i)e^{-tL_N}v_j\rangle\end{aligned} \tag{42}$$

in which the action of the L_N on the v_i to its left was achieved through integration by parts. Differentiating a second time yields

$$\begin{aligned}\frac{d^2}{dt^2}M_{ij}&=\sum_{i<l}\frac{1}{m}\kappa_{il}\langle(q_i-q_l)L_Ne^{-tL_N}v_j\rangle-\sum_{l<i}\frac{1}{m}\kappa_{li}\langle(q_l-q_i)L_Ne^{-tL_N}v_j\rangle\\&=-\sum_{i<l}\frac{1}{m}\kappa_{il}\langle(v_i-v_l)e^{-tL_N}v_j\rangle+\sum_{l<i}\frac{1}{m}\kappa_{li}\langle(v_l-v_i)e^{-tL_N}v_j\rangle\\&=\frac{1}{m}\sum_l\kappa_{il}\langle v_le^{-tL_N}v_j\rangle-\frac{1}{m}\sum_l\kappa_{il}\langle v_ie^{-tL_N}v_j\rangle\\&=\frac{1}{m}\sum_l\kappa_{il}M_{lj}(t)-\frac{1}{m}\sum_l\kappa_{il}M_{ij}(t)\ .\end{aligned} \tag{43}$$

Define Ω_{ij} by

$$\begin{aligned}(\Omega^2)_{ij}&=\begin{bmatrix}\frac{1}{m}\left[\sum_l\kappa_{il}\right], & i=j\\ -\frac{1}{m}\kappa_{ij}, & i\neq j\end{bmatrix}\\&=-\frac{1}{m}\kappa_{ij}+\frac{1}{m}\left[\sum_l\kappa_{il}\right]\delta_{ij}\ .\end{aligned} \tag{44}$$

Therefore, we can now write

$$\frac{d^2}{dt^2}M_{ij}(t)=-\sum_l(\Omega^2)_{il}M_{lj}(t) \tag{45}$$

with the initial conditions

$$M_{ij}(0)=\langle v_iv_j\rangle=\frac{k_BT}{m}\delta_{ij}$$

and (46)

$$\frac{d}{dt}M_{ij}(0)=-\langle v_i L_N v_j\rangle=0 .$$

The solution is simply the matrix

$$M_{ij}(t)=\frac{k_BT}{m}[\cos(t\Omega)]_{ij} . \tag{47}$$

An equivalent result was obtained by Ford, Kac, and Mazur.[46]

So far, the coupling matrix κ_{ij} is completely general. If we put the oscillators in a circle and let them interact with nearest neighbors only, with identical strengths, and write $\omega^2\equiv(1/m)\kappa$, we then have

$$(\Omega^2)_{ij}=-\omega^2(\delta_{i+1,j}+\delta_{i-1,j})+2\omega^2\delta_{ij} . \tag{48}$$

The eigenvectors and eigenvalues for this matrix are known and are easily verified to be

$$\frac{1}{\sqrt{N}}\exp\left[i\frac{2\pi}{N}ls\right]$$

with eigenvalue (49)

$$\lambda_l=4\sin^2\left[\frac{\pi}{N}l\right]\omega^2 .$$

This is to be interpreted as

$$\sum_{s=1}^{N}(\Omega^2)_{js}\frac{1}{\sqrt{N}}\exp\left[i\frac{2\pi}{N}ls\right]=\lambda_l\frac{1}{\sqrt{N}}\exp\left[i\frac{2\pi}{N}ls\right] . \tag{50}$$

It leads to the spectral resolution of $\cos(t\Omega)$ given by

$$[\cos(t\Omega)]_{jk}=\frac{1}{N}\sum_{s=1}^{N}\exp\left[i\frac{2\pi}{N}s(k-j)\right]\times\cos\left[2\omega t\sin\left[\frac{\pi}{N}s\right]\right] . \tag{51}$$

In particular, the velocity autocorrelation for any oscillator is given by a diagonal element of this expression

$$\langle vv(-t)\rangle_N=\frac{k_BT}{mN}\sum_{s=1}^{N}\cos\left[2\omega t\sin\left[\frac{\pi}{N}s\right]\right] . \tag{52}$$

In the limit $N\to\infty$, we find

$$\lim_{N\to\infty}\langle vv(-t)\rangle_N=\frac{k_BT}{m}\int_0^1 dx\cos[2\omega t\sin(\pi x)]=\frac{k_BT}{m}\frac{2}{\pi}\int_0^{\pi/2}dy\cos[2\omega t\sin(y)] \tag{53}$$

$$=\frac{k_BT}{m}J_0(2\omega t) , \tag{54}$$

where J_0 is the Bessel function of order 0. It is interesting that it has the asymptotic form

$$J_0(2\omega t)\sim\frac{\cos\left[t-\frac{\pi}{4}\right]}{t^{1/2}} \tag{55}$$

which reminds one of the long-time tail for one dimension, except for the oscillatory numerator. This numerator is sufficient to produce a finite Green-Kubo integral:

$$D=\int_0^\infty dt\langle vv(-t)\rangle=\frac{k_BT}{m}\int_0^\infty dt\,J_0(2\omega t)=\frac{k_BT}{2m\omega} . \tag{56}$$

The notion that momentum conservation is responsible for long-time tails is incompatible with this result. In two or more dimensions, nearest-neighbor coupling leads to more complicated expressions[45] and $D\equiv0$. That the diffusion constant vanishes makes sense because there cannot be real diffusion in a fixed, oscillating lattice. That $D\neq0$ in one dimension is a kind of fluke. One can only speculate about whether the numerator or the denominator in (55) is indicative of the generic behavior for a fluid system.

The possibility that a superposition of cosines, as in (52), is the form of the velocity autocorrelation function in general, that is, for a fluid system rather than a lattice, arises in the Mori-Zwanzig theory of correlation functions.[47] The continued fraction representation[48,49] for the Laplace transform of the relaxation function (in the present context, the velocity autocorrelation function) takes the form

$$\Xi(Z)=\cfrac{1}{Z+\Delta_1\cfrac{1}{Z+\Delta_2\cfrac{1}{Z+\Delta_3\cfrac{1}{Z+\cdots}}}} . \tag{57}$$

While the theory provides formal expressions for the

Δ_i's, they are not computable in general. If at any stage, a Δ_j is set equal to zero, then $\Xi(Z)$ may be written as a ratio of polynomials in Z which has the form of the Laplace transform of a sum of cosines. These cosines each have different amplitudes and arguments, which depend upon the values of the Δ_i's. In the event that no Δ_j equals zero, we expect $\Xi(Z)$ is a superposition of infinitely many cosines. The spectral density of their arguments and the distributions of amplitudes will determine the nature of the behavior of their sum. Whether long-time tails or much more rapid decays occur is simply not known. Both long-time tails and exponential decays, as well as many other possibilities, are compatible with this representation of the autocorrelation function. When the frequencies are linearly independent much is already known[50] about such series, but it is not known when a physical system will give rise to linearly independent frequencies.

VI. STOKES-BOUSSINESQ

The idea that long-time tails are fundamentally hydrodynamic in origin gains considerable support from a very old result in classical hydrodynamic theory, the Stokes-Boussinesq formula.[14–17] This formula provides time-dependent corrections to the simpler Stokes formula for viscous drag on a sphere in uniform motion. There are two corrections. One is the Archimedean correction to the inertial mass, while the other involves an integral over the past and gives rise to a long-time tail in the velocity autocorrelation function. This long-time tail is identical with that already discussed in the preceding sections.

The Stokes-Boussinesq formula for the drag on a sphere of radius R in a fluid of mass density ρ and viscosity η is

$$M\frac{d}{dt}\vec{v}=-6\pi\eta R\vec{v}-\tfrac{1}{2}M_0\dot{\vec{v}} +6\pi\eta R^2\left[\frac{\rho}{\eta}\right]^{1/2}\int_{-\infty}^{t}ds\frac{\dot{\vec{v}}(s)}{\sqrt{t-s}}\ . \tag{58}$$

In the Introduction, the effect of the first term on the right-hand side of (58), the Stokes friction term, was exhibited in Eqs. (1)—(3). A simple exponentially decaying velocity autocorrelation function (1) was obtained and Einstein's formula (3) for the diffusion constant resulted. Physically, the simple Stokes friction occurs when a sphere is somehow caused to move at constant velocity through a fluid. This is really incompatible with the exponentially decaying velocity implied by the Stokes equation. The nonuniform velocity equation of Stokes-Boussinesq (58) leads to a velocity autocorrelation function given by[14]

$$\langle v(t)v(0)\rangle=\frac{k_BT}{2\pi}\int_C d\omega\frac{e^{-i\omega t}}{-i\omega M+\gamma(\omega)} \tag{59}$$

in which the contour C in the complex ω plane is taken in the clockwise sense along the real axis and then back along a semicircle around the bottom half of the ω plane and $\gamma(\omega)$ is obtained from (58) by Fourier transform and is

$$\gamma(\omega)=6\pi\eta R[1+R(-i\omega/\nu)^{1/2}-i\omega R^2/9\nu]\ , \tag{60}$$

where ν is the kinematic viscosity of the fluid. The square root is defined by a branch cut along the negative real axis in the ω plane. If (59) is used on the left-hand side of (3), then one still gets the right-hand side. Thus, D is insensitive to the long-time tail. It has been shown[14] that the expression in (59) does yield

$$\langle v(t)v(0)\rangle\simeq\frac{2}{3}\frac{k_BT}{\rho}(4\pi\nu|t|)^{-3/2} \tag{61}$$

as $t\to\infty$.

Even though D is insensitive to the long-time tail, the mean-square displacement of the sphere obeys two different laws as $t\to\infty$ depending on whether Stokes friction or Stokes-Boussinesq friction is used to calculate it. This is the basis for the light-scattering experiments of Paul and Pusey.[20] For Stokes friction alone, the $t\to\infty$ behavior of the mean-square displacement goes linearly in t, whereas the Stokes-Boussinesq friction implies an asymptotic $t^{1/2}$ correction. Paul and Pusey clearly saw the difference, although the amplitude of the observed effect was only 74% of the predicted value. Their observations were for polystyrene spheres, 17 000 Å in radius. Several possibilities exist to explain the discrepancy in amplitude, but this issue is not fully settled yet, and is not of direct concern in the context of the present paper. I am inclined to state that Paul and Pusey have seen the Stokes-Boussinesq effect.

In the remainder of this section, I will argue that the Stokes-Boussinesq formula must be properly interpreted before it can be used as a basis for the velocity autocorrelation function for a Brownian particle. I will also argue that the Paul and Pusey observation is of a macroscopic character which cannot simply be extrapolated to a truly microscopic regime for molecules or systems of identical hard spheres.

The difficulty with the Stokes-Boussinesq equation (58) was already alluded to above while discussing the simpler Stokes equation. To derive the

Stokes friction, a *steady*-state hydrodynamics problem is solved. Either a sphere moves at *constant* velocity through an infinite fluid, or an infinite fluid moves at *constant* asymptotic velocity past a fixed sphere. These two perspectives are equivalent and it is useful to take the former perspective since it is much more easily generalized to the Stokes-Boussinesq case. This means that an outside force of some kind keeps the sphere moving at *constant* velocity, and the consequent viscous drag caused by the fluid is computed. This drag determines how much work must be done by the outside force to keep the sphere moving at *constant* velocity. If the outside force is suddenly removed, then it is imagined that the velocity decays according to the exponential decay law implicit in (1). This is in fact not proved, but is merely an extrapolation of the result for steady motion to nonsteady motion. The generalization to nonsteady motion is described in great detail by Landau and Lifshitz.[15] First, they consider what would happen if an outside force is used to make a sphere oscillate with very small amplitude and fixed frequency ω. The time-dependent Navier-Stokes equation is now required. The resulting fluid velocity field and drag force is calculated. The ultimate result is a drag force such as in (58) except that the $\dot{\vec{v}}$'s must be replaced by the corresponding expression for a sphere oscillating at *constant* frequency. In principle, the fluid will react to a forced oscillating sphere by first evolving according to transient solutions to the Navier-Stokes equations and by secondly evolving according to particular, forced solutions. It is only the latter solutions which are used in the derivation of the Stokes-Boussinesq equation. Landau and Lifshitz then proceed to show that if the outside force creates a forced motion which can be expressed by a Fourier integral over fixed frequencies, then the drag force on the sphere induced in the fluid is the Fourier superposition of the responses obtained for each frequency. Again, no transient behavior is included. Landau and Lifshitz actually never wrote an equation such as (58). They have written the drag force on the right-hand side of (58) and they solve problems in which the $\dot{\vec{v}}$ in this expression is *specified from outside.* This gives them the induced drag force caused asymptotically by a sphere which suffers an imposed motion from outside forces. For example, they consider the case in which $\dot{\vec{v}}$ is constant. Based on their argument, there is no reason to expect the result to hold for the *self*-force on a Brownian sphere. That is, one is *not* justified in writing (58) in a situation in which no outside force is contemplated, but in one which it is imagined that the drag force is induced by the motion of the sphere without outside forces. In short, we have not justified the *self*-forcing equation (58) on the basis of the Stokes-Boussinesq drag force calculation, which is for forced motion of the sphere. This is simply the generalization of the objection to writing

$$M\frac{d\vec{v}}{dt} = -6\pi\eta R\vec{v} \tag{62}$$

based on a calculation of Stokes friction determined for a sphere which *must move* at constant velocity. This is not to say that there are not regimes in which either (58) or (62) are good approximations; rather, they are not justified by the standard derivations of their respective drag forces. In the case of (62), justification requires a separation of time scales. Specifically, if the fluid relaxation, governed by the time scale R^2/ν, is short compared with the velocity relaxation time scale $M/(6\pi\eta R)$, then (62) is a good approximation. This amounts to

$$M \gg 6\pi\rho R^3 = \tfrac{9}{2}M_0 \tag{63}$$

in which M is the mass of the Brownian sphere and M_0 is the mass of the displaced fluid [which also appears in the Archimedean term in (58)]. In the experiments of Paul and Pusey, for example, it was found convenient to work with polystyrene spheres in water made up of H_2O-D_2O mixtures. These mixtures were used to obtain $M = M_0$, so that settling effects could be minimized. Thus, they did their work well outside the validity of (62), which was their intention. However, this leaves open the question of whether (58) is valid. This question is much harder to answer since the corresponding calculation which includes transients has not been exhibited.

There exists a parallel problem for which the transient behavior can be deduced and compared with the results which would be the analogue of (58). This is the problem of the drag produced on an infinite plane surface which undergoes lateral motion in its own plane. It is considered by Landau and Lifshitz[51] explicitly, and a mathematically equivalent heat-flow problem is treated by Morse and Feshbach.[52] The problem is a Dirichlet boundary value problem.

Let the plane be the y-z plane. An incompressible viscous fluid is in the region $x > 0$ with the plane at $x = 0$. From symmetry, it is evident that all quantities depend on x and t only. The velocity of the plane is denoted by $\vec{v}$ and the velocity field of the fluid is denoted by $\vec{u}$.[53] We assume that the plane's motion is parallel to the y axis and oscillatory. The boundary conditions for the fluid's velocity field are $\vec{u} = \vec{v}$ at $x = 0$, i.e., $u_x = u_z = 0$ and $u_y = v$. Incompressibility requires $\vec{\nabla}\cdot\vec{u} = 0$, which becomes

$(\partial/\partial x)u_x=0$, which with boundary condition $u_x=0$ at $x=0$ implies $u_x=0$ everywhere. Moreover, $(\vec{u}\cdot\vec{\nabla})\vec{u}$, the nonlinear term in the Navier-Stokes equation, becomes $[u_x(\partial/\partial x)]\vec{u}$ since there can be x-t dependence only, and this vanishes because $u_x=0$. The equation of motion for the fluid becomes

$$\frac{\partial}{\partial t}\vec{u}=-\frac{1}{\rho}\vec{\nabla}p+\nu\nabla^2\vec{u}\ . \tag{64}$$

The x component of (64) is $\partial p/\partial x=0$ which implies that the pressure p is constant since it has no y-z dependence. Thus, letting u_y be denoted by simply u yields

$$\frac{\partial}{\partial t}u=\nu\frac{\partial^2}{\partial x^2}u \quad \text{with} \quad u=v \quad \text{at} \quad x=0\ , \tag{65}$$

a classical Dirichlet problem, and it is equivalent, mathematically, with a heat conduction problem[52] with the temperature given at $x=0$. The Dirichlet Green's function is readily obtained by the method of images and is

$$G_D(x,x',t,t')=4\pi\nu(4\pi\nu|t-t'|)^{-1/2}\left[\exp\left[-\frac{(x-x')^2}{4\nu|t-t'|}\right]-\exp\left[-\frac{(x+x')^2}{4\nu|t-t'|}\right]\right] \tag{66}$$

which has a normal derivative at $x'=0$ given by

$$-\frac{\partial}{\partial x'}G_D(x,0,t,t')=-\frac{4\pi x}{|t-t'|}(4\pi\nu|t-t'|)^{-1/2}\times\exp\left[-\frac{x^2}{4\nu|t-t'|}\right]\ . \tag{67}$$

Incidentally, we again see a long-time tail emerging from the *normalization* factor of the *Gaussian* solution to a *diffusion* equation. The solution can be written in general as

$$u(x,t)=\frac{1}{4\pi}\int_0^t dt'\frac{\partial}{\partial x'}G_D(x,0,t,t')u(0,t')+\frac{1}{4\pi\nu}\int_0^\infty dx'G_D(x,x',t,0)u(x',0) \tag{68}$$

in which $u(0,t')$ is, of course, $v(t')$, and we will take $u(x',0)=0$. Therefore, the solution is

$$u(x,t)=\int_0^t dt'\frac{x}{2\sqrt{\pi\nu}}\frac{\exp\left[-\frac{x^2}{4\nu|t-t'|}\right]}{|t-t'|^{3/2}}v(t')\ . \tag{69}$$

Morse and Feshbach[52] state the following: "In order to obtain a steady-state solution, we shift the 'beginning of time' to $t=-\infty$ at which time initial values are zero." Landau and Lifshitz do the same without saying so. The idea is to eliminate transients. For the case of a pure frequency oscillation, $v(t')=v_0\cos(\omega t')$ and we get

$$u_0(x,t)=\int_0^t dt'\frac{x}{2\sqrt{\pi\nu}}\frac{\exp\left[-\frac{x^2}{4\nu|t-t'|}\right]}{|t-t'|^{3/2}}\times v_0\cos(\omega t') \tag{70}$$

from (69), or

$$u_\infty(x,t)=\int_{-\infty}^t dt'\frac{x}{2\sqrt{\pi\nu}}\frac{\exp\left[-\frac{x^2}{4\nu|t-t'|}\right]}{|t-t'|^{3/2}}\times v_0\cos(\omega t') \tag{71}$$

from the shift of the time origin. Morse and Feshbach[52] find

$$u_\infty(x,t)=v_0\exp\left[-\left[\frac{\omega}{2\nu}\right]^{1/2}x\right]\cos\left[\omega t-\left[\frac{\omega}{2\nu}\right]^{1/2}x\right]$$

$$=v_0\mathrm{Re}\left\{\exp\left[i\omega t-\left[\frac{i\omega}{\nu}\right]^{1/2}\right]\right\}, \tag{72}$$

where Re means real part. By a related calculation, it is found that

$$u_0(x,t)=v_0\mathrm{Re}\left\{\exp\left[i\omega t-\left[\frac{i\omega}{\nu}\right]^{1/2}x\right]\right\}-\frac{v_0}{\pi}\,\mathrm{Im}\left\{\int_0^\infty dy\,\frac{y\,e^{-yt}}{y^2+\omega^2}\exp\left[i\left[\frac{y}{\nu}\right]^{1/2}x\right]\right\} \tag{73}$$

in which Im means imaginary part. To obtain this result, complex integration was required in which all square roots are interpreted by a branch cut along the negative real axis.

The drag force experienced by the plane is given by the stress tensor

$$\sigma_{xy}=\eta\frac{\partial u}{\partial x}\bigg|_{x=0^+}. \tag{74}$$

This is, in fact, the force *per unit area* on the plane caused by the fluid viscosity. Landau and Lifshitz

$$u_\infty(x,t)=\frac{1}{2\pi}\mathrm{Re}\left\{\int_{-\infty}^{\infty}v(\omega)\exp\left[i\omega t-\left[\frac{i\omega}{\nu}\right]^{1/2}x\right]d\omega\right\}. \tag{76}$$

This leads to a force per unit area given by

$$\sigma_{xy}=\left[\frac{\rho\eta}{\pi}\right]^{1/2}\int_{-\infty}^{t}\frac{dv(s)}{ds}\frac{1}{\sqrt{t-s}}ds \tag{77}$$

very similar to the Stokes-Boussinesq expression. It is evident that a different result is obtained if the analogue of (73) is used instead.

The essential point of the preceding discussion is that it would not be justified *a priori* to write the *self*-forcing equation

$$\mu\frac{d}{dt}v=\eta\frac{\partial u}{\partial x}\bigg|_{x=0^+}, \tag{78}$$

where μ is the mass per unit area of the plane. Certainly, the transient behavior in (73) should not be ignored. Here, the plane experiences the force it engenders through the fluid viscosity and it is not subject to any outside forcing. Equation (78) would be potentially valid only in some appropriate regime in which one would think of the plane's relaxation taking place on a much slower time scale from the time scale during which the fluid relaxes to its *steady-state* response as given by (76) and (77). From (73) the transient contribution to (74) can be computed and is

$$\sigma_{xy}^{\mathrm{trans}}=-\rho v_0\frac{\sqrt{\nu}}{\pi}\int_0^\infty dy\,\frac{y^{3/2}e^{-yt}}{y^2+\omega^2}. \tag{79}$$

In fact, this expression goes asymptotically like $t^{-5/2}$, i.e., like

$$\sigma_{xy}^{\mathrm{trans}}\underset{t\to\infty}{\to}-\rho v_0\frac{\sqrt{\nu}}{\pi}\frac{\Gamma(\frac{5}{2})}{\omega^2 t^{5/2}}. \tag{80}$$

In this case, the asymptotic regime begins only after $t\gg\omega^{-1}$.

The self-force will be sensitive to the immediate past. The expression given by (77) is the drag force are plainly interested in the effect of forcing the plane to oscillate at a fixed frequency by some external means. Consequently, it makes sense to wait until transients die out, and to use (72) in (74) rather than (73). They also consider the case where $v(t)$ is not purely oscillatory but is a Fourier superposition of frequencies:

$$v(t)=\frac{1}{2\pi}\mathrm{Re}\left[\int_{-\infty}^{\infty}v(\omega)e^{-i\omega t}d\omega\right] \tag{75}$$

with the interpretation attending (71). They then get

which affects a plane forced from outside in a prescribed manner. It ignores the immediate past including the entire transient. By taking the initial time at $t=-\infty$ it is designed to ignore the transient. In the case of the sphere, it would be of interest to work out in detail the transient response. Even so, one should not expect the result to significantly modify the conclusions based upon the Stokes-Boussinesq equation. This is so because the transient behavior will exhibit a long-time—tail behavior characteristic of diffusion equations, of which the Navier-Stokes equation is a special kind. Moreover, diffusion equations are well known to be especially poor descriptions of short-time behavior.[54] Consequently, it would not be expected that a complete analysis including transients would yield a good result because the crucial behavior during the immediate past is poorly described. The self-forcing problem poses special difficulties. In the regime of molecular Brownian motion, the rate of thermal perturbations is such that viscoelastic effects must also be contemplated. These effects result from frequency dependence in the viscosity, η, for sufficiently high frequencies.[55,56] For molecular Brownian motion in water, the inclusion of such effects are necessary, but they render the problem of a sphere in a viscoelastic Navier-Stokes fluid extremely difficult. By itself, the viscoelastic correction does not remove the general concern regarding the poor short-time description of diffusion equations.

Even given all these caveats, the Stokes-Boussinesq equation for self-forcing, Eq. (58), probably has a regime of validity just as does the simpler Stokes equation for self-forcing, Eq. (62). We do not know yet how to delimit this regime. If we accept the observations of Paul and Pusey at face value, then we know that the regime is *not* characterized in the same way as the regime for (62) was characterized, i.e., by (63), as was discussed earlier.

In any case, I believe that the measurements worked out as they did because the polystyrene spheres were so "large" (17 000-Å radius). The issue here has to do with temperature fluctuations and the dependence of viscosity on temperature. The qualitative argument is that for a large sphere, the temperature fluctuations in a volume of comparable size are negligible, whereas for a sphere of molecular size, the temperature fluctuations are so big that the temperature dependence of the viscosity destroys the idea of a *constant* viscosity on the molecular scale. This renders the hydrodynamic description moot, if not invalid, at the molecular level. Quantitatively, the temperature fluctuations in a volume V are given by

$$\langle \Delta T_V \Delta T_V \rangle = \frac{k_B T^2}{C_V} \tag{81}$$

in which T is the average temperature and C_V is the heat capacity of the volume V. If we take $T=300$ K and use the specific heat for water, which is $c=1$ cal/K mole, we find

$$\langle \Delta T_V \Delta T_V \rangle = 5.3\times 10^{-18}(\mathrm{K})^2 \frac{\mathrm{cm}^3}{V} \tag{82}$$

in which V is to be measured in cm^3. For the polystyrene spheres, $V=2\times 10^{-11}$ cm^3. Thus, the root-mean-square temperature fluctuations are of order

$$\langle \Delta T_V \Delta T_V \rangle^{1/2} = 5\times 10^{-4}\ \mathrm{K}\ . \tag{83}$$

In contrast, a molecule of radius 10 Å (a small protein) has a volume $V=4.2\times 10^{-21}$ cm, which yields root-mean-square temperature fluctuations of order

$$\langle \Delta T_V \Delta T_V \rangle^{1/2} = 36\ \mathrm{K}\ . \tag{84}$$

Coupling this fact with the fact that for water[57]

$$\frac{\partial \ln \eta}{\partial \ln T} \simeq -2\ , \tag{85}$$

which means that the viscosity changes noticeably with temperature, implies that the viscosity will lose its meaning in the neighborhood of the molecule. This seems to be a more serious problem than viscoelasticity or the poor, short-time description of diffusion.

All of the preceding considerations lead to the conclusion that there is no justified connection between the computer simulations of the velocity autocorrelation function for hard spheres (and the theories purporting to explain the same) and the Stokes-Boussinesq equation (and the apparent observations of the same). In this regard, the last sentence of Sec. 6.1 of Chap. XII of the book by Résibois and DeLeener[6] is of interest: "The real surprise is that these macroscopic arguments also apply to the long-time behavior of correlation functions for microscopic particles." Do they?

VII. LORENTZ MODEL

The Lorentz-gas model was developed because of the impossibility of solving a realistic N-body dynamics problem. In this model, a single particle moves in a space (usually the two-dimensional Euclidean space R^2) of fixed scatterers with constant speed. When contact with a scatterer occurs, a simple, elastic reflection takes place, changing the direction of the velocity. A variety of systematic and stochastic variations of the model are reviewed by van Beijeren.[26]

The Lorentz model was developed by Lorentz[58] to describe conduction electrons in metals. Its popularity in kinetic theory stems from its nontrivial nature and tractability. It is not to be confused with the Lorenz model,[21] with which is associated the idea of a strange attractor.

The velocity autocorrelation function for the Lorentz model has been shown[26] to possess a long-time tails. This has been taken as support for the kinetic-theory approach[18] to long-time tails. It could, however, be argued that it shows that the Lorentz model provides a good modeling for hydrodynamic, or diffusive, correlations. In light of the discussion earlier in this paper, this need not be construed as evidence for long-time tails in exact dynamics.

There is another objection of a more profound nature. The work of Bunimovich and Sinai[25] suggests that the long-time–tail results may not be generic. They introduced the concept of a finite horizon, which is not featured in the traditional accounts of the model. A Lorentz model has finite horizon[25] if there is a constant A such that the length of any straight segment (of the path of the scattering particle) which avoids all scatterers cannot be more than this constant A. They proved the theorem[25] which says the following:

There exists a constant γ, $0<\gamma\leq 1$, such that for all sufficiently large n

$$|E(v(n),v(0))| \leq \exp(-n^{\gamma})\ . \tag{86}$$

In this expression, n is the number of collisions which have occurred, $v(n)$ is the velocity after the nth collision, $v(0)$ is the initial velocity, and $|E(v(n),v(0))|$ is essentially what we have been denoting by $\langle v(t)v(0)\rangle$. If $\gamma<1$, the decay law is not quite an ordinary damped exponential, but it is far more similar to one than to a long-time–tail reciprocal power law.

The approach of Bunimovich and Sinai is so dif-

ferent from the traditional kinetic-theory approach that the apparent conflict between these two approaches has not yet been resolved. The idea of a finite horizon seems to be intuitively clear. It suggests the transition from a Lorentz "gas" to a Lorentz "liquid." Can one incorporate the idea of a finite horizon into the kinetic-theory approach and corroborate the result obtained by Bunimovich and Sinai?

VIII. DYNAMICAL SYSTEMS

The expression "dynamical system" has come to mean different things to physicists and mathematicians. For the physicist, it usually means a system described by a Hamiltonian with some definite interaction potentials. For the mathematician, it has come to mean some kind of differentiable manifold with a flow or mapping defined on it. The manifold is further characterized either topologically, geometrically, by a measure, or by some combination of these. The physicist usually studies the particular properties of his specific system, whereas the mathematician is looking for generic properties in his. The interface between these two approaches is especially difficult, and except for certain simple cases, it is not known which specific Hamiltonians fall into which generic categories. Consequently, an application of the mathematical view of dynamical systems to the question of the velocity autocorrelation function is not yet possible in a definitive fashion.

In Sec. II one type of application was discussed. It involved the application of the Anosov-Bowen theorem to the question of numerical error propagation in computer simulations. As was pointed out in Sec. II, even this application is fraught with subtleties.

Another aspect of the difficulty in making contact between the two disciplines of the physicist and the mathematician in the context of the present paper is that a great deal of the mathematical work on dynamical systems has been for dissipative systems rather than for Hamiltonian systems. This is especially true of much of the work on attractors and on bifurcations.[21,59] Of the work on Hamiltonian systems, much of it has been on perturbation questions for systems with few particles, such as in celestial mechanics or with coupled oscillators. The truly remarkable Kolmogorov-Arnold-Moser (KAM) theorem[60] is a product of these endeavors.

The problem of the velocity autocorrelation function for a tagged particle in an N-particle system has not been the object of intense investigation from the mathematical viewpoint. With the exception of the work on the Lorentz gas mentioned in Sec. VII, the context for studies of correlations has been the full phase space. In this context the central concept[21,32] is that of "hyperbolicity." Both Axiom-A systems and Anosov systems are defined in terms of this concept. The essence of the idea may be expressed as follows:

Let f^t be a diffeomorphism on a compact manifold M. Let Λ be a closed invariant submanifold of M, i.e., $\Lambda \in M$ and $f^t\Lambda = \Lambda$. Let v_x^t be in the tangent bundle at the point x of Λ with respect to f^t. The tangent bundle over Λ is denoted by $T_\Lambda M$. Λ is said to be hyperbolic with respect to f^t if there is a continuous splitting of $T_\Lambda M$ in $E^0+E^s+E^u$, where E^0 is a one-dimensional subspace along the flow, E^s is a contracting subspace (s for stable) such that

$$||v_x^t|| \leq c\, e^{-\lambda t}||v_x^0||$$

for $t>0$ and $x \in E^s$, and E^u is a dilating subspace (u for unstable) such that

$$||v_x^t|| \geq c\, e^{\lambda t}||v_x^0||$$

for $t>0$ and $x \in E^u$, where c and λ are constants, and the double bars denote an appropriate metric.

If the whole of M is hyperbolic with respect to f^t, then f^t is an Anosov flow. It is the nature of the dilating subspace which causes initially close trajectories to exponentially move apart in such systems. One would like to say more, although this is not at all proved; e.g., the projection of such a flow down onto a single particle's velocity will result in a velocity autocorrelation possessing rapid (exponential) decay. Maybe "physical" systems are not Anosov. In fact, as was pointed out in Sec. II, hard spheres are not Anosov.[27] Nevertheless, the presence of intersphere potentials could make such systems Anosov after all. These questions need to be pursued.

Studies of systems with one or a few particles (spheres or disks) in specially bounded regions (usually of R^2) suggest another variation on this theme.[61] A dynamical system may have islands of stability in a sea of hyperbolicity. The islands of stability contribute long-time tails to the correlation function whereas the hyperbolic sea leads to an exponential decay. The relative contributions, i.e., the amplitudes, depend upon the measures of these two subspaces of the whole manifold. There is some reason to believe that as the number of particles increases, the measure of the stable islands decreases exponentially. This tantalizing suggestion needs more study. There is the possibility that it is related with the computer simulations. The connection involves the size of the box used in the simulations

and the density of disks. If both are increased, perhaps islands of stability, which would be responsible for the long-time–tail observations, will become much less significant.

IX. CONCLUSION

The existence of long-time tails in the velocity autocorrelation function for a tagged particle in an N-particle system has been questioned. Computer simulations have been criticized with regard to potential numerical error propagation artifacts. The invocation of the β-shadow theorem (Anosov-Bowen theorem) as a defense against this criticism has also been questioned. Three theoretical approaches have also been criticized or questioned. Their mutual agreement was shown to result from the fact that they each ultimately utilize a hydrodynamic correlation mechanism. Thus, they do not really provide independent corroboration of each other. Moreover, it was argued that they may merely describe the correlations resulting from numerical error propagation which were heuristically shown to be potentially of hydrodynamic character. Nevertheless, the kinetic-theory approach is impressive. The experimental observation of long-time tails was analyzed from the viewpoint of the Stokes-Boussinesq effect. It was argued that this is perhaps a purely "macroscopic" effect, with no justified correlate at the microscopic level. Finally, it was argued that the approach of the mathematical theory of dynamical systems suggests in several ways that the velocity autocorrelation function of a particle decays much more rapidly than long-time tails imply. The Lorentz-gas model was cited, as well as work on billiard (spheres and disks) systems.

Our objective in this paper is not to deny the existence of long-time tails for the velocity autocorrelation function, or to even imply that some of the evidence is in error somehow. Rather, the objective is to raise a variety of questions which do not seem to be easily answered and which need to be answered before a firm basis for long-time tails is established. Because so much of the evidence is of hydrodynamic character, one suspects that there has been a complacency about the evidence; not because there is so much independent corroboration, but because people have mistakenly viewed the evidence as independent. A truly independent line of enquiry based upon the theory of Anosov systems (or an appropriate variant) does exist, and it is desirable to determine its degree of consistency with the other approaches.

If the numerical error propagation problem could be pushed back as far as, for example, $50t_0$ without altering the presently observed emergence of the long-time–tail behavior around $12t_0$, then this would provide convincing evidence. It may require waiting for the next generation of computers to be achievable. If, on the other hand, the onset of the long-time tail would also be pushed back to $50t_0$ by this procedure, then convincing evidence for the artifact interpretation would seem to be in hand. Work has been done which compares the greater accuracy of 120-bit (binary digit) arithmetic to 60-bit arithmetic. If numerical error is the source of the "hydrodynamiclike behavior" manifested in the long-time tail, then the long-time–tail behavior should show itself in the 60-bit study before it appears in the 120-bit study. It may already be possible to make a definitive statement about this on the basis of existing data.

Although "real" experimental evidence has been recently published, its interpretation has been seriously questioned in Sec. VI of this paper. Consequently, we feel that it is not yet inappropriate to close with a remark made by Ruell[21]:

"In fact one should beware of the general tendency of theoreticians to provide explanations of phenomena which have actually never been observed."

ACKNOWLEDGMENTS

This work was supported by National Science Foundation Grant No. PHY-79-21541. Informative discussions with J. Erpenbeck, J. Ford, B. Holian, T. Keyes, and F. Vivaldi were very helpful. (This does not imply that any of these people endorse the views expressed in this paper.)

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