RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 49, NUMBER 5 MAY 1994

Fluctuating hydrodynamics of the classical electron gas

P.-M. Binder
Wolfson College, Oxford OX2 6UD, United Kingdom
and Center for Nonlinear Studies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 18 November 1993)

Fluctuations of the diffusion tensor in a lattice Lorentz gas model are calculated numerically from a sample of mesoscopic-size systems, and used as input in a mode-coupling expression that predicts long-time tail amplitudes. The results are in good agreement with previously published direct measurements of the tail amplitude over a wide range of scatterer concentrations. We argue that the previous, long-standing disagreement between theory and simulations comes from concentration-independent configurational fluctuations in the diffusion tensor, which we have measured separately and which are not taken into account by existing theories.

PACS number(s): 51.10.+y, 05.60.+w, 66.30.−h

The Lorentz model [1] was proposed originally to study the electrical conductivity of solids, and as a simple model for transport theory. It consists of one or more mutually noninteracting classical particles which collide with fixed, randomly placed scatterers. It has been an important test case in the development of kinetic theory: the derivation of macroscopic equations, generalized kinetic equations, the calculation of transport coefficients, and the study of long-time tails of self-correlation functions [2–9]. It is also the first and one of the most important models for diffusion in a static random medium. The asymptotic behavior of its velocity autocorrelation function [VACF or $\phi(t)$], related to the diffusion coefficient by the Green-Kubo formula, has puzzled kinetic theorists for over two decades. Only very recently has agreement been obtained between theory [3] and simulations [10–12] for the *exponent* of the algebraic long-time tails (t^{-2} in two dimensions) of the VACF for general scatterer concentrations.

In the present paper we consider a fluctuating hydrodynamics (FH) semiphenomenological mode-coupling theory developed by Ernst *et al.* [13]. The results of this theory, discussed below, relate fluctuations of the diffusion tensor $K_{\alpha\beta}$ to long-time tail amplitudes. We find good agreement between previously measured [10] long-time tail amplitudes and FH predictions which use as input our measurements of the diffusion tensor fluctuations. By performing simulations of a lattice model with either strictly constant or fluctuating concentration, we can isolate concentration-induced and configuration-induced contributions to the long-time tails. We conclude that for the lower scatterer concentrations, configurational effects (i.e., concentration-independent fluctuations) account for previous discrepancies between theory and simulation (see Refs. [3–8] and the discussion two paragraphs below).

The FH theory of Ernst *et al.* begins with the fundamental assumption that the detailed microscopic description of the random medium (number and positions of scatterers) can be replaced by a more macroscopic description, in terms of a spatially varying diffusion tensor and a quantity related to free volume. Based on this a coarse-grained fluctuating diffusion equation was developed, and a general expression was calculated relating long-time tail amplitudes a in the VACF firstly to the Fourier modes of the spatially varying quantities just described, and finally to the variance of $K_{\alpha\beta}$ about its average value as follows:

$$\phi(t) \sim -a/t^2 = -\Delta_K/8\pi t^2, \tag{1}$$

where

$$\Delta_K = \frac{\left\langle \sum_{\alpha,\beta} (K_{\alpha\beta} - \langle K_{\alpha\beta}\rangle)^2 \right\rangle}{2V(D\psi)^2}. \tag{2}$$

Here α, β are the Cartesian coordinates, D is the diffusion coefficient, V is the volume of the subsystems over which $K_{\alpha\beta}$ is measured, and ψ is the volume fraction available to diffusing particles. The brackets denote an average over mesoscopic subsystems of size V of a larger macroscopic system. For the purposes of simulation, it was essential to decouple the mesoscopic subsystems in order to ensure that particles stayed within a given one, so that truly *local* diffusion coefficients could be measured [14]. The main justification for doing this is that (2) no longer contains spatial information about the system, such as dependence on Fourier modes.

We will use $\Delta K_{\alpha\beta}$ to denote the numerator of (2), which is the variance of the diffusion tensor. For the two-dimensional (2D) Lorentz gas, the authors of Ref. [13] specifically substituted for $\Delta K_{\alpha\beta}$ the effect of concentration fluctuations obtained from statistical considerations:

$$\Delta K_{\alpha\beta} = 2\left(\frac{dD}{dc}\right)^2 \mathrm{Var}(c), \tag{3}$$

where $\mathrm{Var}(c) = \bar{c}(1-\bar{c})/V$ is the variance in c. Here $\bar{c}$ denotes the average scatterer concentration of the fluctuating ensemble. When this expression, only valid as $c \to 0$, is substituted into (2), the result agrees with the predictions of the earlier kinetic mode-coupling theory of Ernst and Weyland [3]. At higher concentrations, the approximation of the diffusion tensor fluctuations by concentration-induced fluctuations fails badly. In a recent report of very accurate simulations of the continuum

Lorentz gas, Lowe and Masters [12] found values of the tail amplitude that were up to six times larger than the prediction based on this approximation.

We now proceed to describe the model being studied, and the numerical technique employed to perform accurate measurements of the diffusion tensor of mesoscopic systems with volume V. Such phenomenological data had not been measured before for the 2D Lorentz gas, and therefore the more general results of FH [Eqs. (1) and (2)] have remained untested.

We consider a model in which independent particles move ballistically with unit speed along the principal directions of the 2D triangular lattice, and collide isotropically with point scatterers, which occupy the lattice nodes random and independently [15, 16] with probability c. Long-time tail amplitudes and coefficients for this model have been reported in the literature [10], and are known with uncertainty of about 10% (see Table I). In the present model $\psi = 1$ as all scatterer sites are accessible to the moving particles. Because of lattice symmetries $\langle K_{\alpha\beta}\rangle = D\delta_{\alpha\beta}$.

The measurement of the diffusion tensor, as required in Eqs. (1) and (2) was performed with the moment propagation method which is known to yield excellent statistics. The method consists in evolving distributions of particles, rather than individual particles, according to the microscopic rules of the model. The method averages over all trajectories consistent with a given scatterer configuration. It is described in detail in Refs. [10,11]. In contrast with Ref. [10], in which only the K_{xx} component was considered, here we measure the full diffusion tensor. We have done this by measuring all possible coefficients $K'_{\gamma\epsilon} = \sum_{t=0}^{t=T}\langle v_\gamma(t)v_\epsilon(0)\rangle - 1/4\delta_{\gamma\epsilon}$, where γ, ϵ are the principal directions of the lattice. The above are Green-Kubo expressions, in which the additional constant term, known as the "propagation" term, is due to the discreteness of time in the model; it is discussed by Binder and Ernst [16] and references therein. Summing the projections of each coefficient on the α, β Cartesian coordinates yields the diffusion tensor $K_{\alpha\beta}$. We stress that this measurement is *independent* of that of long-time tails, as we integrate the velocity correlations $\langle v_\gamma(t)v_\epsilon(0)\rangle$. In fact, the long-time tails only produce very small corrections to the diffusion tensor itself, and Eqs. (1) and (2) apply even if $K'_{\gamma\epsilon}$ is measured for times shorter than the onset of long-time tails; this was not done because of the loss of accuracy in the measurement of $K_{\alpha\beta}$.

In order to measure fluctuations of the diffusion tensor, systems of size $V = L^2$, $L = 72$, were simulated over $T = 400$ time steps, which corresponds to 80–360 mean free times for the concentrations considered [17]. L was chosen to be large enough for $K_{\alpha\beta}$ to be meaningful, and small enough for it to fluctuate strongly. The number of independent scatterer configurations ranged from 40 for the denser systems to 1200 for $c = 0.2$, where the slower convergence of the average fluctuation required more sampling. Scatterer concentrations were allowed to fluctuate, by virtue of placing scatterers with independent probability c at each site. A few systems of sizes $L = 36$ and $L = 144$, used as checks, yielded results consistent with those obtained for $L = 72$.

A summary of results is given in Table I. Column 1 shows the values of c for which long-time tail amplitudes are available in the literature for this model (Ref. [10]); the measured amplitude values are reproduced in column 2. Column 3 shows the FH prediction for amplitudes from Eqs. (1) and (2), with D and $\Delta K_{\alpha\beta}$ coming from the simulations just described. Column 4 shows the contributions to column 3 from concentration fluctuations in the mesoscopic systems, following (3). D, Var(c), and dD/dc were obtained directly from simulations. The theoretical values for this quantity, given in parentheses, in column 4, are given by (3) with $D(c)$ and dD/dc coming from the expression $D(c) = (5/12c) - (1/6)$ from Ref. [18], which takes into account backscattering trajectories, and $\mathrm{Var}(c) = \bar{c}(1-\bar{c})/V$ as before. We see that for $c = 0.8$, 0.9, the total fluctuations in the diffusion tensor are well approximated by the concentration-induced ones alone, but at lower concentrations this approximation yields values that are up to 35% low. We tested concentration-independent effects separately for the lower densities, by measuring the variance of the diffusion tensor for 1200 systems of size $L = 80$ with the scatterer concentration fixed at exactly c. The results are given in column 5, which added to the theoretical value from column 4, yield the improved values of column 6, which we consider the true predictions of the FH theory for the three lowest densities. Moreover, this separate test confirms the effects of concentration-independent fluctuations in the model. These values,

TABLE I. First column: scatterer concentration. Second column: direct measurement of long-time tail amplitudes, from Ref. [10]. Third column: fluctuating hydrodynamics predictions, with diffusion coefficient and diffusion tensor variances measured numerically. Fourth column: numerical (and analytical) estimates of fluctuating concentration-induced long time tails. Fifth column (lower concentrations): contribution to long-time tails from fixed-density ensemble (configurational effects). Sixth column: sum of the contributions from column 4 (theory) and column 5 (configurational). Last column: numerically measured diffusion coefficient, given for completeness.

c	a_{num}	a_{FH}	a_{conc} (Theory)	a_{conf}	a_{tot}	D_{num}
0.2	0.463	0.453	0.327 (0.325)	0.130	0.455	1.924
0.4	0.178	0.214	0.172 (0.147)	0.045	0.192	0.868
0.6	0.091	0.094	0.091 (0.080)	0.016	0.096	0.520
0.8	0.038	0.039	0.038 (0.037)			0.351
0.9	0.018	0.016	0.017 (0.019)			0.294

along with the results of column 3 for the two higher densities, should be compared with the amplitudes of column 2. The agreement to about 10% over such a wide range of densities is remarkable.

The error bars in the numerical measurements of columns 3 and 5 were estimated as follows. Each sample (typically 1200 points) was divided into 10 subsets, a small but meaningful number. Average fluctuations were calculated for each subset, and then the standard deviation of the 10 average fluctuations was calculated. The result was always between 10% and 15% of the value of the overall average fluctuation, which we therefore consider a good estimate of the uncertainty in the values reported.

In summary, we have shown that a fluctuation-based theory predicts the amplitude of long-time tails of the Lorentz gas to within 10% of the values reported in the literature. These results are consistent with the error bars in the fluctuations of the diffusion tensor (typically 10–15 %) which the theory requires as input. Our results lend support to the derivation of Eqs. (1) and (2). The fact that predicted amplitudes fall equally above and below simulations suggests that the discrepancies are statistical rather than systematic. From Table I one sees that at low concentrations the contribution from concentration-independent fluctuations in the diffusion tensor accounts for about one third of the value of the long-time tails. We do not presently understand why the discrepancy between measurements and predictions based on concentration-induced theories is much smaller in lattice Lorentz models than in the continuum Lorentz models. We suspect it may have to do with the much wider spectrum of free paths between collisions and also with the existence of trapping areas in the continuum model. We expect the findings of this paper to be relevant to other lattice models of diffusion in random media. We also hope that recent experimental work [19] on gas mixtures which has confirmed some of the predictions of the high-density kinetic theory of Lorentz gases may soon be extended to consider long-time tails.

The author would like to thank the Institute for Theoretical Physics at Utrecht for its hospitality, M.H. Ernst and D. Frenkel for useful discussions, and particularly H. van Beijeren for his participation during much of this work. This work was partly supported by Wolfson College and by the U. S. Department of Energy.

[1] H. A. Lorentz, Proc. K. Ned. Akad. Wet. **7**, 438, (1905); **7** 585 (1905); **7**, 684 (1905); P. Ehrenfest, *Collected Scientific Papers* (North-Holland, Amsterdam, 1959), p. 229.

[2] J. M. J. van Leeuwen and A. Weyland, Physica **36**, 456 (1967); A. Weyland and J. M. J. van Leeuwen, *ibid.* **38**, 35 (1968).

[3] M. H. Ernst and A. Weyland, Phys. Lett. **34A**, 39 (1971).

[4] C. Bruin, Phys. Rev. Lett. **29**, 1670 (1972); Physica **72**, 261 (1974).

[5] B. J. Alder and W. E. Alley, J. Stat. Phys. **19**, 341 (1978).

[6] J. C. Lewis and J. A. Tjon, Phys. Lett. **66A**, 349 (1978).

[7] W. Götze, E. Leutheusser, and S. Yip, Phys. Rev. A **23**, 2634 (1981); **24**, 100 (1981).

[8] T. Keyes and J. Mercer, Physica A **95**, 473 (1979); A. Masters and T. Keyes, Phys. Rev. A **26**, 2129 (1982).

[9] For recent work see P. M. Bleher, J. Stat. Phys. **67**, 461 (1992); L. A. Bunimovich and S. E. Troubetzkoy, *ibid.* **67**, 289 (1992); A. S. Cukrowski, Chem. Phys. **159**, 39 (1992); J. Kortus and C. Olesky, J. Phys. A: Math. Gen. **25**, 1093 (1992); H. Sumi, Solid State Commun. **85**, 1 (1993); A. Baranyai, D. J. Evans, and E. G. D. Cohen, J. Stat. Phys. **70**, 1085 (1993), and references therein.

[10] P.-M. Binder and D. Frenkel, Phys. Rev. A **42**, 2463 (1990).

[11] D. Frenkel, F. van Luijn, and P.-M. Binder, Europhys. Lett. **20**, 7 (1992).

[12] C. P. Lowe and A. J. Masters, Physica A **195**, 149 (1993).

[13] M. H. Ernst, J. Machta, J. R. Dorfman, and H. van Beijeren, J. Stat. Phys. **34**, 477 (1984); J. Machta, M. H. Ernst, H. van Beijeren, and J. R. Dorfman, *ibid.* **35**, 413 (1984).

[14] We have excluded scatterer configurations allowing infinite free paths, as these would give rise to an infinite diffusion tensor; such configurations have zero probability of occurring in infinite systems. We have also checked our configurations, generated with the Berkeley random number generator, for correlations such as those found by A. M. Ferrenberg, D. P. Landau, and Y. J. Wong, Phys. Rev. Lett. **69**, 3382 (1992). We found that the scatterer to scatterer distance, n, followed closely the expected distribution, $\mathrm{Prob}(n) \sim c(1-c)^n$, and therefore that no serious correlation effects appear in our systems. Finally, we note that the spectrum of decay rates of simple diffusive modes becomes discrete in this case and as a consequence Eqs. (1) and (2) will hold but only up to a cutoff time $t \sim L^2 D^{-1}$, where L is the system size: see L. F. Perondi and P.-M. Binder, Phys. Rev. B **48**, 4136 (1993). Finally, the finite-size corrections for D in a system of size L are of order $(1/L^2)$, and therefore are negligible for the system sizes considered here: see M. J. A. M. Brummelhuis and H. J. Hilhorst, J. Stat. Phys. **53**, 249 (1988) and L. F. Perondi and P.-M. Binder, Phys. Rev. B **47**, 14 221 (1993).

[15] M. H. Ernst and P.-M. Binder, J. Stat. Phys. **51**, 981 (1988).

[16] We have chosen the triangular lattice to avoid the effects of staggered invariants, as described in Ref. [9] and in P.-M. Binder and M. H. Ernst, Physica A **164**, 91 (1990).

[17] We estimate the error e from truncating the simulation after 400 time steps as follows: since the long-time tails have already set in Ref. [10], the neglected contribution to the diffusion tensor measurement is of order $a\int_{400}^{\infty} t^{-2}dt \sim a/400$. As $a < D$, $e/D < 1\%$.

[18] H. van Beijeren and M. H. Ernst, J. Stat. Phys. **70**, 793 (1993).

[19] R. Bongratz and Ch. Morkel, J. Non-Cryst. Sol. **156**, 205 (1993).