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Breakdown of the Boltzmann equation in cellular-automata lattice gases

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In lattice gases the Boltzmann equation is not valid at low densities, if the collision rules admit reflections of unlike particles because of long-lived correlated ring-type collisions. This is shown for a simple mixture, a Lorentz gas, by comparing theoretical and molecular dynamics results for the diffusion coefficient.

Simulations of cellular automata (CA) models for two-dimensional nonequilibrium fluids[1] seem to yield results for hydrodynamic and transport phenomena that are very well described by the nonlinear Boltzmann equation which only accounts for uncorrelated collisions. This is true not only at low and high densities (because of particle-hole symmetry), but, surprisingly, also at intermediate densities, where at most small deviations from Boltzmann occur. It holds for the viscosity,[1] as well as for tagged particle diffusion[2] both in CA games with deterministic or stochastic collision rules. Similar quantitative agreement has been found in lattice versions of Lorentz gases.[3,4]

To investigate this puzzle one needs to develop a systematic kinetic theory for CA fluids that enables one to calculate systematically higher-order density corrections to the Boltzmann equation. We further want to avoid the difficulties, inherent to two-dimensional fluid-type models, in which transport coefficients are proportional to the logarithm of the system size. In order to do so we have chosen a simple CA fluid, namely a lattice Lorentz gas, which consists of fixed scatterers on a fraction c of the N sites (chosen at random) of a square lattice with unit lattice distance. *Independent* particles move at integer times $t=0,1,2,\ldots$ from one site to its nearest-neighbor site along straight lines in any of the four lattice directions, $\mathbf{e}_\nu$ ($\nu=1,2,3,4$ [mod(4)]) and are scattered upon collision with scatterers. Examples of deterministic scattering rules are given in Refs. 3 and 4.

Here we choose stochastic collision rules, defined through the three probabilities α, β, and γ with the normalization $\alpha+\beta+2\gamma=1$, where α is the transmission probability, β the reflection probability, and γ the deflection probability in an orthogonal direction. By varying these parameters one may possibly fine tune to certain pathological features of the lattice models; backscattering ($\beta\neq 0$) is one of them.

It is well known in the literature[5,6] that the possibility of backscattering and retracing trajectories may create long-time memory effects that can change the (low density) Boltzmann value of the diffusion coefficient by a substantial fraction [as is the case in a one-dimensional (1D) gas of hard rods (Ref. 5)] or it can even make the diffusion coefficient vanish [as is the case in the Ehrenfest wind-tree model with overlapping trees (Ref. 6)]. Similar things are happening in lattice-gas models.

To illustrate this we will compare the phase space of all possible scatterer positions, compatible with the collision sequences in Figs. 1(a)-1(d). This will be done in the limit of small density of scatterers c and of large times t where t is typically on the order of the mean free time $t_{\rm mf}\sim 1/c$. In the case of uncorrelated collisions [see Fig. 1(a)], which are accounted for in the Boltzmann equation, the phase space for k uncorrelated collisions is proportional to $(ct)^k\sim O(1)$. In a similar way one estimates the phase space of ring collisions [see Fig. 1(b)] and orbiting collisions (as defined in Ref. 6) to be $c(ct)^{k-1}\sim O(c)$; the phase space for retracing "nested" ring collisions with backscattering [see Fig. 1(c)] is the *same* as for the uncorrelated collisions; without backscattering [see Fig. 1(d)] one has $c(ct)^{k-1}\sim O(c)$.

These estimates show the breakdown of the Boltzmann equation for models *with backscattering* ($\beta\neq 0$). Here, at low densities the contributions to transport coefficients and time correlation functions of the correlated collision sequences in Fig. 1(c), for times on the order of several mean free times, are of equal importance as the uncorrelated collisions of Fig. 1(a). The Boltzmann equation only sums these uncorrelated collision sequences. In order to calculate the leading low-density behavior the correlated collisions sequences of $O(1)$ have to be summed as well.

In models *without backscattering* ($\beta=0$) the Boltz-

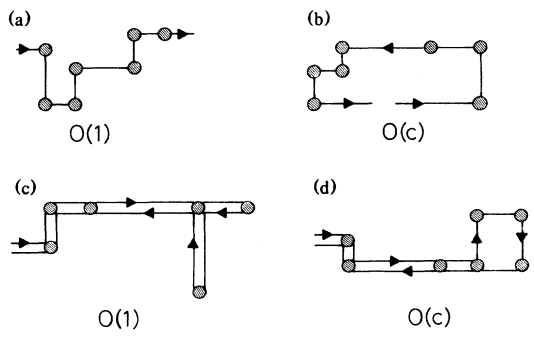

FIG. 1. Collision sequences: (a) uncorrelated, (b) simple rings, and nested rings (c) with and (d) without reflections.

mann equation does take the dominant low-density contributions into account. To calculate the first density correction of relative $O(c)$ one has to sum not only the ring collisions of Fig. 1(b), but also the orbiting collisions and the nested ring collisions of Fig. 1(d).

After these phase-space arguments we outline our quantitative calculations. A more detailed analysis will be published elsewhere.[7] We start from the Chapman-Kolmogorov or stochastic Liouville equation for the probability distribution of finding a particle at time t *arriving* at site $\mathbf{n}$ with velocity v (i.e., coming from site $\mathbf{n}-\mathbf{e}_v$) in a given configuration of scatterers $\{c_{\mathbf{n}}\}$. With each site $\mathbf{n}$ a random variable $c_{\mathbf{n}}$ is associated that assumes the values 0 or 1 with probability $1-c$ or c, respectively, corresponding to the absence or presence of a scatterer at site $\mathbf{n}$. We use a $4N\times 4N$ matrix notation with labels $(\mathbf{n}\nu,\mathbf{m}\mu)$ and write the Liouville equation for the matrix of conditional probabilities as

$$P(t+1)=S^{-1}(1+CT)P(t)\,,$$

with $P(0)=1$ and $P(1)=S^{-1}$. Here S is the translation; C is the fluctuating density of scatterers with $C_{\mathbf{n}\nu,\mathbf{m}\mu}=c_{\mathbf{n}}\delta_{\mathbf{nm}}\delta_{\nu\mu}$ and T is the collision operator with $T_{\mathbf{n}\nu,\mathbf{m}\mu}=\delta_{\mathbf{nm}}T_{\nu\mu}$, where

$$T_{\nu\mu}=(\alpha-1)\delta_{\nu\mu}+\beta\delta_{\nu\mu+2}+\gamma(\delta_{\nu\mu+1}+\delta_{\nu\mu-1})\,.$$

To obtain the average probabilities the matrix $P(t)$ (or its generating function) must be averaged over all configurations of scatterers $\{c_{\mathbf{n}}\}$,

$$\sum_{t=1}^{\infty}\xi^t\langle P(t)\rangle=\langle[(1+z)S-1-CT]^{-1}\rangle$$

with $\xi=1/(1+z)$. The quantities of main interest in this article are the diffusion coefficient D and the velocity autocorrelation function (VACF) $\phi(t)$, defined as

$$\phi(t)=(4N)^{-1}\sum_{\mathbf{n}\nu,\mathbf{m}\mu}e_{\nu x}e_{\mu x}\langle P_{\mathbf{n}\nu,\mathbf{m}\mu}(t)\rangle\,.$$

The average depends only on $\mathbf{n}-\mathbf{m}$ because of translational invariance. The brackets represent an average over the quenched variables $\{c_{\mathbf{n}}\}$. The diffusion coefficient is given by

$$D=\tfrac{1}{4}+\sum_{t=1}^{\infty}\phi(t)\,.$$

We start our kinetic theory analysis by considering the Boltzmann approximation, which sums all uncorrelated collision sequences (in which a moving particle never returns to the same scatterer). This approximation is obtained from the Liouville equation by replacing the matrix of fluctuating densities of scatterers C by its average $\langle C\rangle=c$. Then the subsequent equations yield the well-known Boltzmann results[4,8] $\phi_B(t)=\frac{1}{2}(1-c\tau_1)^t$ and $D_B=(2c\tau_1)^{-1}-\frac{1}{4}$ with $\tau_1=1-\alpha+\beta$. In Fig. 2 the Boltzmann value D_B is shown by dashed lines (for $\gamma=\frac{1}{2}$ dashed and solid lines coincide). These results are exact for $c=1$, where our model reduces to a random walk on a uniform lattice. We note that D_B contains a term $-\frac{1}{4}$, which is of relative $O(c)$ as $c\to 0$.

To study correlated collision sequences we perform a

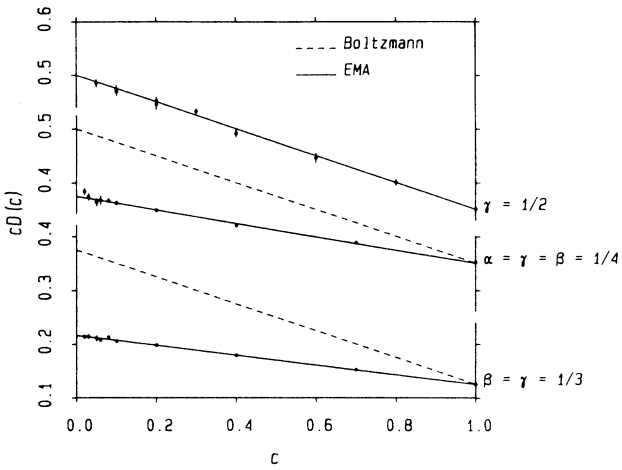

FIG. 2. Diffusion coefficient vs density of scatterers. Dashed and solid lines show theoretical results from the Boltzmann equation and effective medium approximation; dots with error bars show results of molecular dynamics simulations. Model $\{\gamma=\frac{1}{2},\ \alpha=\beta=0\}$ has no reflections (Boltzmann and effective medium approximation curves coincide). Models $\{\alpha=\beta=\gamma=\frac{1}{4}\}$ and $\{\alpha=0,\ \beta=\gamma=\frac{1}{3}\}$ have reflections.

formal perturbation expansion of $\langle P(t)\rangle$ in powers of the fluctuation $\delta C=C-c$, using standard methods of kinetic theory. The terms in the perturbation expansion are analyzed in the limit as $z\to 0$ (corresponding to $t\to\infty$). One finds that the ring, nested ring, and orbiting collision terms are dominant and yield the estimates given at the start of this article with ct replaced by c/z. In the low-density limit the nested ring collisions are resummed by the following self-consistent equation for the 4×4 ring matrix $R(z)$, which is similar in structure to the self-consistent mode coupling equations[1]

$$R(z)=\int_q\{(1+z)e^{i\mathbf{q}\cdot\mathbf{V}}-1-c[T+TR(z)T]\}^{-1}\,,$$

with the 4×4 matrix $(V_a)_{\mu\nu}=e_{\nu a}\delta_{\mu\nu}$ with $a=x,y$. The symbol $\int_q$ represents a volume average over the first Brillouin zone. If $R(z)$ on the right-hand side is set equal to zero, this equation reduces to the *simple* ring integral, which has for small z and $|\mathbf{q}|$ a structure identical to the continuous Lorentz gas. The diffusion coefficient that follows from there as $c\to 0$ is

$$D=(2c\tau_1)^{-1}(1+r_1\tau_1)-\tfrac{1}{4}\,,$$

where r_1 is an eigenvalue of the ring matrix $R(0)$. In the limit of $z\to 0$ and small c the eigenvalues of $R(0)$ can be determined analytically. Two essentially different cases have to be distinguished: *Without reflections* ($\beta=0$) the relevant eigenvalue is found to be $r_1=cr_0(1-2\gamma)/(1-3\gamma/2)$, where $r_0=-(1/\pi-\frac{1}{4})\gamma$ is the corresponding eigenvalue of the simple ring matrix. *With reflections* ($\beta\neq 0$) the relevant eigenvalue is $r_1=-\frac{1}{2}+a/[1+(1+8ab)^{1/2}]$ with $a=(\alpha+\gamma)/(\beta+\gamma)$ and $b=(\alpha+\beta)/(2\gamma)$.

Some comments on the low-density results are in order: Without reflections the (negative) r_1 gives an $O(c)$ correction to cD_B in the last equation. If r_1 is replaced by

r_0 one includes only the $O(c)$ terms from simple ring collisions [see Fig. 1(b)]. It is also remarkable that in the special model with only right and left turns ($\gamma=\frac{1}{2}$; $\alpha=\beta=0$) the eigenvalue r_0 of the simple ring is nonvanishing, but the corresponding eigenvalue r_1 of the summed (nested) rings is vanishing (ring terms are canceled by nested rings). In general, the correction $|r_1/2c|$ in the equation for D is very small (less than ≈ 0.01). The $O(c)$ term is not exact because we have neglected the orbiting events, mentioned in the introduction.

The models *with reflection* yield very different results. Here the contribution r_1, originating from the nested ring collisions, is seen to be *independent of the density* as $c\rightarrow 0$, in agreement with the estimates at the start of this article. The value of D is 25 to 50% smaller than D_B as $c\rightarrow 0$ (depending on the model parameters α,β,γ), as can be seen in Fig. 2 by comparing dashed and solid lines for $\gamma\neq\frac{1}{2}$. This demonstrates the breakdown of the Boltzmann equation as a valid low-density kinetic theory in models with reflection. However, the diffusion coefficient $D_{NR}(c)$, calculated from the last equation for the nested ring collisions, does *not* represent the *exact* value of $cD(c)$ as $c\rightarrow 0$. In fact, all treelike collision trajectories [as in Fig. 1(c)], in which the branches are retraced an arbitrary number of times, are proportional to $(ct)^k\sim O(1)$ for $t\rightarrow\infty$ and $c\rightarrow 0$, similar to those in Figs. 1(a) and 1(c). We have not yet been able to sum all treelike collision trajectories and determine the exact value of $cD(c)$ at $c=0+$ for models with reflections.

In the high-density limit, at a low concentration of "holes" (empty sites), one can apply similar perturbation techniques as for small c, and use $p=1-c$ as a small parameter.

For intermediate densities we have developed[7] an effective medium approximation (EMA), in which the matrix $T+TR(z)T$ in the equation for $R(z)$ is replaced by an effective medium collision matrix $T_{\rm eff}(z)$. The EMA equations can be solved numerically and yield the results shown as solid (almost straight) lines in Fig. 2. In the low-density limit the EMA equations reduce to those for the nested ring collisions.

The above results can also be compared with computer simulations. Ruijgrok has performed preliminary molecular dynamics (MD) simulations using an ATARI ST1040 for the models: $\gamma=\frac{1}{2}$, $\alpha=\beta=0$, and $\alpha=\beta=\gamma=\frac{1}{4}$ using lattices of 360×360 and up to 200 time steps. Simulation data are obtained by averaging over 10 configurations of scatterers and over 2500 particle trajectories per configuration. Preliminary runs for models with reflections were performed at Centre Européen de Calcul Atomique et Moléculaire, University of Orsay. The data points shown are obtained on a SUN3-160 using typically 40 configurations of scatterers on lattices up to 500×500 sites and typically 2000 particle trajectories (of up to 1500 time steps) per configuration. The data are shown in Fig. 2. There is very good agreement between the simulation and the EMA results at all densities. The present theory also offers an explanation of the simulation results for the diffusion coefficient, obtained by Ruijgrok and Cohen[3(b)] for the lattice Lorentz gas with a fifty-fifty mixture of fixed mirrors, scattering under angles of plus or minus $\pi/2$. If the total concentration of scatterers is not too high, and the time not too long, their model is expected to correspond to our stochastic model with left-right scattering only.

What are the implications of the above analysis for CA models of nonequilibrium fluids? The concept of backscattering or reflection is meaningless in single-component CA fluid of indistinguishable unlabeled particles as in the Hardy, de Pazzis, and Pomeau (HPP) model or the Frisch, Hasslacher, and Pomeau (FHP) model.[1] However, it is a natural collision rule to be allowed in collisions between unlike particles in CA mixtures. The phase space for ring-type collisions with reflections at low densities and long times is again of the same magnitude as that of the uncorrelated collisions. Consequently, we expect a breakdown of the standard nonlinear Boltzmann equation with Fermi exclusion as a valid kinetic equation at low densities. If reflections are not allowed, we expect that the Boltzmann equation is correct at low densities.

It would be of interest to test these expectations by computer simulations on the simple example of mutual diffusion in a spatially inhomogeneous mixture of red and blue particles, that are otherwise identical. If one disregards the color, the mixture is in thermal equilibrium. The simulations can be performed on the usual FHP models, if one assigns an extra Boolean color variable to the link $\mathbf{e}_\nu$ at node $\mathbf{n}$, if the link is occupied.

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