Generalized Boltzmann Equation for Lattice Gas Automata

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In this paper a theory is formulated that predicts velocity and spatial correlations between occupation numbers that occur in lattice gas automata violating semi-detailed balance. Starting from a coupled BBGKY hierarchy for the n-particle distribution functions, cluster expansion techniques are used to derive approximate kinetic equations. In zeroth approximation the standard nonlinear Boltzmann equation is obtained; the next approximation yields the ring kinetic equation, similar to that for hard-sphere systems, describing the time evolution of pair correlations. The ring equation is solved to determine the (nonvanishing) pair correlation functions in equilibrium for two models that violate semi-detailed balance. One is a model of interacting random walkers on a line, the other one is a two-dimensional fluid-type model on a triangular lattice. The numerical predictions agree very well with computer simulations.

1. INTRODUCTION

The most important generalization of Boltzmann's kinetic equation is the so-called ring equation,[1] which accounts for correlated collision sequences. In the kinetic theory of classical fluids[2,3] one expands the two-, three-, etc., particle distribution functions or time correlation functions systematically in cluster or correlation functions by neglecting all higher-order correlations except pair correlations. In this manner one obtains the nonlinear generalized Boltzmann equation for the single-particle distribution function and the ring equation for the pair correlation function. In

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the context of lattice gas automata (LGA) it was first used in 1991 by Kirkpatrick and Ernst[4] following the standard methods for continuous systems.

The kinetic theory studies of the 1960s[1-3] showed that these techniques can be applied equally well to the distribution functions and time correlation functions; they can be applied directly to the Liouville operators, or to the BBGKY hierarchy[2] satisfied by distribution functions or time correlation functions.

These standard methods of kinetic theory have been used to obtain the ring kinetic theory for time correlation functions in LGA, either for fluid-type models[4,5] or for purely diffusive models.[6-8] In particular, ring contributions to transport coefficients have been evaluated numerically for FHP models by Brito and Ernst[9] and compared with computer simulations by Gerits *et al.*[10],3 Another successful application of ring kinetic theory to LGA is the derivation of mode coupling theory and long-time tails.[4]

Very recently,4 Boghosian and Taylor[11] applied a similar method of cluster expansions to the BBGKY hierarchy for LGA by linearizing the hierarchy around the known Gibbsian equilibrium state. These authors characterized the formal structure of the nth-order collision term in the generalized Boltzmann equation. Their work should be considered as the LGA analog of the early studies by Cohen[2] and Green and Picirelli[12] for continuous fluids.

The theories discussed above refer to LGA that obey the so-called semi-detailed balance condition, and consequently approach for long times a Gibbsian equilibrium state, where all distribution functions are known. The method of ref. 11 also applies to models that violate semi-detailed balance only weakly, so that equilibrium properties are not affected, but only nonequilibrium properties such as transport coefficients. Most existing LGA fluids without detailed balance are not of this type.[13-16]

The calculation of the correlation functions in equilibrium for models that *strongly* violate detailed balance is the main goal of this paper. We apply cluster expansion techniques to the BBGKY hierarchy of multiparticle distribution functions. The resulting *nonlinear* generalized Boltzzmann equation and ring kinetic equation can be solved to obtain the single-particle and pair distribution functions in the unknown *non-Gibbsian* equilibrium state.

We describe the problems of interest in the current paper in somewhat more detail. In the theory of lattice gas automata (LGA) the Gibbs distribution is known to be the unique equilibrium distribution if the

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3 Ref. 10 compares the theory of ref. 5 with simulations of 2D triangular LGA.

4 A preprint only became available at the time when the present research was being submitted.

dynamics satisfy the so-called semi-detailed balance[17] or Stueckelberg[18] condition. The Gibbs distribution depends on the phase space variables *only* through globally conserved quantities, such as the total number of particles N, the total momentum $\mathbf{P}$, or (for thermal models) the total energy H. If the collision rules are strictly local, involving particles on the same node only, then the Gibbs equilibrium state is completely factorized, i.e., the occupations of velocity channels at the same or at different nodes are completely uncorrelated.

However, in LGA that violate the condition of semi-detailed balance equal-time correlations are know to exist in equilibrium between occupation numbers of different velocity channels on the same or on different nodes.[13,16,19] The existence of such equilibrium correlations prevents the equilibrium distribution from being the Gibbs distribution, and therefore standard equilibrium statistical mechanics does not apply to LGA that violate (semi-) detailed balance. In the literature a quantitative or qualitative understanding of the structure of the equilibrium distribution for such models is entirely lacking. Fundamental questions are: Is there a unique equilibrium state, or can there be several stationary states? To what extent does the final state depend on the initial state and on the details of the collision rules? How does the system approach the correlated equilibrium state? How can one calculate transport properties and time correlation functions? These questions are partly answered in the present paper.

The organization of the paper is as follows. In Section 2 we derive the first two equations of a coupled hierarchy for the joint distribution functions, and we introduce a cluster expansion around mean-field theory that leads to a closure of the hierarchy at the level of two-point correlation functions. In Section 3 the coupled kinetic equations are specialized to spatially uniform stationary states, and equations are constructed for the stationary values of the single-particle distribution function and the pair correlation function; an iterative scheme is discussed for evaluating these equations numerically. To establish the accuracy of the theory, in Section 5 we apply the theory to a model of interacting random walkers on a line and to a fluid-type model on the triangular lattice. We compare the theoretical predictions with detailed computer simulations performed for both models. We end with some conclusions and remarks about future developments in Section 6.

2. KINETIC THEORY

2.1. Microdynamic Equation

We consider a lattice gas automaton defined on a regular d-dimensional lattice $\mathscr{L}$ with periodic boundary conditions, containing $V = L^d$ nodes. On

each node $r \in \mathscr{L}$ there exist b allowed velocity channels $\mathbf{c}_1, \mathbf{c}_2,..., \mathbf{c}_b$, corresponding to the nearest-neighbor lattice vectors; there may also be rest particles with $|\mathbf{c}_i| = 0$. The system evolves at discrete time steps $t = 0, 1,...$. The microscopic configuration of the system at time t is given in terms of occupation numbers $n_i(\mathbf{r}, t) = \{0, 1\}$, denoting the absence or presence of a particle in velocity channel $(\mathbf{r}, \mathbf{c}_i)$. The state of node $\mathbf{r}$ is denoted by $n(\mathbf{r}, t) = \{n_i(\mathbf{r}, t); i = 1, 2,..., b\}$.

One time step in the evolution involves an instantaneous *collision* step which transforms a precollision state $n(\mathbf{r}, t)$ into a postcollision state $n^*(\mathbf{r}, t)$ on all nodes $\mathbf{r}$ independently. It is followed by a *propagation* step,

$$n_i(\mathbf{r} + \mathbf{c}_i, t + 1) = n_i^*(\mathbf{r}, t) \tag{2.1}$$

during which a particle at node $\mathbf{r}$ is moved to the nearest-neighbor node $\mathbf{r} + \mathbf{c}_i$ in the direction of its velocity $\mathbf{c}_i$. The collision step is specified in terms of a matrix of *transition probabilities* $A_{s\sigma}$ from a precollision or in-state $s = \{s_1, s_2,..., s_b\}$ to a postcollision or out-state $\sigma = \{\sigma_1, \sigma_2,..., \sigma_b\}$. The matrix $A_{s\sigma}$ satisfies the *normalization* condition

$$\sum_\sigma A_{s\sigma} = 1 \tag{2.2}$$

There are additional constraints imposed on the transition matrix $A_{s\sigma}$ due to the presence of local conservation laws, which are most easily expressed in terms of the *collisional invariants* $a_i = \{1, \mathbf{c}_i, \frac{1}{2}c_i^2,...\}$, as

$$\sum_i a_i n_i(\mathbf{r}, t) = \sum_i a_i n_i^*(\mathbf{r}, t) \tag{2.3}$$

The local conservation laws imply global conservation of the total quantities $\mathscr{H} = \{N, \mathbf{P}, H,...\} \equiv \sum_{r,i} a_i n_i(\mathbf{r}, t)$. In diffusive LGA only the total number of particles N is conserved; in fluid-type LGA the total momentum $\mathbf{P}$ is conserved in addition. There also exist thermal models where the total energy H is conserved. Furthermore, the majority of LGA have additional (spurious) global invariants,[20] which are mostly "staggered" in space and time. They can be included in the present discussion in a similar manner.

2.2. Semi-Detailed Balance

Much is known about the properties of LGA that satisfy the condition of *semi-detailed balance* with respect to the Gibbs distribution,

$$\sum_s A_{s\sigma} = 1 \tag{2.4}$$

also known as the Stueckelberg condition,[18] or the stronger condition of *detailed balance* with respect to the Gibbs distribution,

$$A_{s\sigma} = A_{\sigma s} \tag{2.5}$$

The latter implies the semi-detailed balance condition (2.4) on account of the normalization condition (2.2). If condition (2.4) is satisfied, then the equilibrium distribution for the model is the Gibbs distribution, which only depends on the globally conserved quantities $\mathscr{H}$.[5]

A comment on the difference between semi-detailed balance and detailed balance is appropriate. We group all states $s(\mathbf{r})$ that can be mapped into one another by lattice symmetries (rotations, reflections) into the same *equivalence class*.[21,14] Next, suppose that for any given in-state $s(\mathbf{r})$, transitions are allowed to out-states $\sigma(\mathbf{r})$ belonging to at most M different equivalence classes. As long as $M \leqslant 2$ there is no distinction between semi-detailed balance and detailed balance. Almost all LGA used in the literature have $M \leqslant 2$. Notable exceptions are the FCHC models.[15]

In what follows, no condition apart from normalization will be imposed on the transition matrix $A_{s\sigma}$. Of course, all the results that will be obtained in the remainder of this paper will hold for LGA that satisfy semi-detailed balance as well, being just a special case of the more general class of LGA violating semi-detailed balance.

2.3. Hierarchy Equations

To construct the equations of motion for the occupation numbers we observe that the occupation numbers $n(\mathbf{r}, t)$ and $n^*(\mathbf{r}, t)$ are related through the transition matrix $A_{s\sigma}$ as

$$n_i^*(\mathbf{r}, t) = \sum_{\sigma s} \sigma_i \hat{A}_{s\sigma} \delta(s, n(\mathbf{r}, t)) \tag{2.6}$$

where $\delta(s, n) = \prod_j \delta(s_j, n_j)$ is a product of b Kronecker delta functions. Similarly, for the product of two occupation numbers on the *same* node we have

$$n_i^*(\mathbf{r}, t) \, n_j^*(\mathbf{r}, t) = \sum_{\sigma s} \sigma_i \sigma_j \hat{A}_{s\sigma} \delta(s, n(\mathbf{r}, t)) \tag{2.7}$$

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[5] The semi-detailed balance and detailed balance conditions with respect to an arbitrary phase space distribution function $P_0(s)$ have respectively the forms $\sum_s P_0(s) A_{s\sigma} = P_0(\sigma)$ and $P_0(s) A_{s\sigma} = A_{\sigma s} P_0(\sigma)$. The phase space distribution in the Gibbs state depends on the variables $s(\mathbf{r})$ only through locally conserved quantities. Consequently $P_0(s) = P_0(\sigma)$ and the (semi-) detailed balance conditions reduce to the equations in the text.

For every node $\mathbf{r}$ and every time t the Boolean realization $\hat{A}_{s\sigma} = \hat{A}_{s\sigma}(\mathbf{r}, t) = \{0, 1\}$ of the $(s \rightarrow \sigma)$ transition is drawn from an $(\mathbf{r}, t)$-independent probability distribution with expectation value $\langle\!\langle \hat{A}_{s\sigma} \rangle\!\rangle = A_{s\sigma}$.

The hierarchy equations describe the time dependence of the distribution functions, i.e., the expectation value of products of occupation numbers. The single-particle and two-particle distribution functions,

$$f_i(\mathbf{r}, t) = \langle n_i(\mathbf{r}, t) \rangle$$
$$f_{ij}^{(2)}(\mathbf{r}, \mathbf{r}', t) = \langle n_i(\mathbf{r}, t) \, n_j(\mathbf{r}', t) \rangle \tag{2.8}$$

are defined as averages $\langle \cdots \rangle$ over an arbitrary distribution of initial occupation numbers, $\{n(\mathbf{r}, 0)\}$. Note that $f_{ii}^{(2)}(\mathbf{r}, \mathbf{r}, t) = f_i(\mathbf{r}, t)$.

We now introduce the single-node and two-node distribution functions,

$$p(s, \mathbf{r}, t) = \langle \delta(s, n(\mathbf{r}, t)) \rangle$$
$$p^{(2)}(s, \mathbf{r}, s', \mathbf{r}', t) = \langle \delta(s, n(\mathbf{r}, t)) \, \delta(s', n(\mathbf{r}', t)) \rangle \tag{2.9}$$

By taking the average of (2.6) over an arbitrary distribution of initial states $\{n_i(\mathbf{r}, 0)\}$ as well as over the $\hat{A}$ distribution of realizations of the transition matrix and using (2.1), we obtain

$$f_i(\mathbf{r} + \mathbf{c}_i, t + 1) = \langle n_i^*(\mathbf{r}, t) \rangle$$

$$= \sum_{\sigma s} \sigma_i A_{s\sigma} \, p(s, \mathbf{r}, t)$$

$$= f_i(\mathbf{r}, t) + \sum_{s\sigma} \delta\sigma_i A_{s\sigma} \, p(s, \mathbf{r}, t) \tag{2.10}$$

where we have used the relation $\delta\sigma_i = \sigma_i - f_i$ and the normalization (2.2). To obtain an analogous equation for the pair distribution function we must distinguish between the cases $\mathbf{r} = \mathbf{r}'$ and $\mathbf{r} \neq \mathbf{r}'$. When $\mathbf{r} \neq \mathbf{r}'$ the collisions for channels $(\mathbf{r}, \mathbf{c}_i)$ and $(\mathbf{r}', \mathbf{c}_j)$ are independent and given by (2.6). When $\mathbf{r} = \mathbf{r}'$, however, we must use (2.7). The combined result can be written as

$$f_{ij}^{(2)}(\mathbf{r} + \mathbf{c}_i, \mathbf{r}' + \mathbf{c}_j, t + 1)$$

$$= \langle n_i^*(\mathbf{r}, t) \, n_j^*(\mathbf{r}', t) \rangle$$

$$= [1 - \delta(\mathbf{r}, \mathbf{r}')] \left\{ \sum_{\sigma s, \sigma' s'} \sigma_i \sigma_j' A_{s\sigma} A_{s'\sigma'} \, p^{(2)}(s, \mathbf{r}, s', \mathbf{r}', t) \right\}$$

$$+ \delta(\mathbf{r}, \mathbf{r}') \left\{ \sum_{\sigma s} \sigma_i \sigma_j A_{s\sigma} \, p(s, \mathbf{r}, t) \right\} \tag{2.11}$$

Equations (2.10) and (2.11) constitute respectively the first two equations of an open hierarchy of coupled equations for the distribution functions, similar to the BBGKY hierarchy for continuous systems. It is straightforward to extend this procedure to construct the hierarchy equations for $f^{(l)}$ with $l \geqslant 3$, although the complexity increases rapidly. In this paper the first two equations of the BBGKY hierarchy suffice for our purpose.

The node distribution functions in (2.11) are related to the l-particle distribution functions in a simple manner. This follows from the following identity, valid for delta functions $\delta(s, n)$ with binary variables as their arguments:

$$\delta(s, n) \equiv \prod_j \delta(s_j, n_j) = \prod_j n_j^{s_j}(1 - n_j)^{1 - s_j} \qquad (2.12)$$

which can be verified directly. As s_j takes only values 0 or 1, the right-hand side is a sum of products of at most b occupation numbers. The average of (2.12), $p(s, \mathbf{r}, t)$, is therefore a linear combination of l-particle distribution functions $f^{(l)}$ with $l = 1, 2,..., b$, all referring to the same node, and with coefficients that are either $+1$ or -1. Similarly $p^{(2)}$ is a linear combination of distribution functions.

To understand the differences between the BBGKY hierarchy for discrete LGA and continuous systems we note the following. In continuous systems usually only two-body interactions (additive forces) are considered; consequently the right-hand side of the lth hierarchy equation contains only $(l + 1)$-particle distribution functions. If one would include s-body interactions $(s = 2, 3, 4,..., b)$ the lth hierarchy equation would contain $(l + b - 1)$-particle distribution functions. In the LGA case b-body interactions simultaneously occur on each node $\mathbf{r}$, where b is the number of velocity channels at a node. Therefore the first hierarchy equation already involves up to b-particle distribution functions; the second equation even involves up to $2b$-particle functions.

2.4. Cluster Expansion

An approximate closure of the hierarchy equations can be obtained by making a cluster expansion in terms of two-, three-, etc., point correlation functions and retaining correlation functions up to a certain order while neglecting the higher-order ones. This procedure leads to approximate kinetic equations.[22]

The cluster functions G are defined through the so-called Ursell expansion,

$$f_{ij}^{(2)}(\mathbf{r}, \mathbf{r}') = f_i(\mathbf{r})f_j(\mathbf{r}') + G_{ij}(\mathbf{r}, \mathbf{r}')$$

$$f_{ijl}^{(3)}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') = f_i(\mathbf{r})f_j(\mathbf{r}')f_l(\mathbf{r}'') + f_i(\mathbf{r})\,G_{jl}(\mathbf{r}', \mathbf{r}'') + f_j(\mathbf{r}')\,G_{il}(\mathbf{r}, \mathbf{r}'')$$

$$+ f_l(\mathbf{r}'')\,G_{ij}(\mathbf{r}, \mathbf{r}') + G_{ijl}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') \tag{2.13}$$

etc., where in general all channels $(\mathbf{r}, c_i)$, $(\mathbf{r}', c_j)$, and $(\mathbf{r}'', c_l)$ are different. Solution of the recursion relations yields the pair and triplet functions in terms of the $f^{(1)}$. The pair and triplet cluster functions can conveniently be written in terms of fluctuations,

$$\delta n_i(\mathbf{r}, t) = n_i(\mathbf{r}, t) - f_i(\mathbf{r}, t) \tag{2.14}$$

For the two-point function we have

$$G_{ij}(\mathbf{r}, \mathbf{r}', t) = f_{ij}^{(2)}(\mathbf{r}, \mathbf{r}', t) - f_i(\mathbf{r}, t)f_j(\mathbf{r}', t) = \langle \delta n_i(\mathbf{r}, t)\,\delta n_j(\mathbf{r}', t) \rangle \tag{2.15}$$

and similarly

$$G_{ijl}(\mathbf{r}, \mathbf{r}', \mathbf{r}'', t) = \langle \delta n_i(\mathbf{r}, t)\,\delta n_j(\mathbf{r}', t)\,\delta n_l(\mathbf{r}'', t) \rangle$$

The expressions are more complicated for four-point and higher-order correlation functions. It is important to note that the diagonal elements for $(\mathbf{r}, c_i) = (\mathbf{r}', c_j)$ are completely determined by $f_i(\mathbf{r}, t)$, due to the Boolean character of $n_i(\mathbf{r}, t)$,

$$G_{ii}(\mathbf{r}, \mathbf{r}, t) = \langle (\delta n_i(\mathbf{r}, t))^2 \rangle = f_i(\mathbf{r}, t)[1 - f_i(\mathbf{r}, t)] \equiv g_i(\mathbf{r}, t) \tag{2.16}$$

To perform the cluster expansion of the hierarchy equations the cluster expansion of the node distribution functions $p(s)$ and $p^{(2)}(s, s')$ is needed. In Appendix A the expansion up to terms linear in the pair correlations is worked out detail. It amounts to obtaining approximate expressions for p and $p^{(2)}$ in terms of f and G. For the single-node distribution function the result is $[\delta s_k = s_k - f_k$ as in (2.14)]

$$p(s, \mathbf{r}, t) = F(s, \mathbf{r}, t) \left\{ 1 + \sum_{k<l} \frac{\delta s_k(\mathbf{r}, t)\,\delta s_l(\mathbf{r}, t)}{g_k(\mathbf{r}, t)\,g_l(\mathbf{r}, t)} G_{kl}(\mathbf{r}, \mathbf{r}, t) + \cdots \right\} \tag{2.17}$$

Here $F(s, \mathbf{r}, t)$ is the *completely factorized* single-note distribution function,

$$F(s, \mathbf{r}, t) = \prod_j [f_j(\mathbf{r}, t)]^{s_j} [1 - f_j(\mathbf{r}, t)]^{1 - s_j} \tag{2.18}$$

In a similar way we can expand the two-node distribution function. Substitution of these expansions in the hierarchy equations yields coupled approximate equations of motion for $f_i(\mathbf{r}, t)$ and $G_{ij}(\mathbf{r}, \mathbf{r}', t)$. For more details we refer to Appendix A.

2.5. Nonlinear Boltzmann Equation

The kinetic equation in *zeroth* approximation is obtained by neglecting G_{kl} in (2.17), or equivalently by replacing the distribution functions $p(s, \mathbf{r}, t)$ in (2.10) by the factorized $F(s, \mathbf{r}, t)$. In that case the first hierarchy equation reduces to the *nonlinear Boltzmann equation*,

$$f_i(\mathbf{r} + \mathbf{c}_i, t + 1) - f_i(\mathbf{r}, t) = \Omega_i^{(1,0)}(f(\mathbf{r}, t)) \qquad (2.19)$$

with the nonlinear collision operator $\Omega_i^{(1,0)}(f)$ given by

$$\Omega_i^{(1,0)}(f) = \sum_{s\sigma} \delta\sigma_i A_{s\sigma} F(s) = \sum_{s\sigma} (\sigma_i - s_i) A_{s\sigma} F(s) \qquad (2.20)$$

The second equality follows from (2.2) and (2.18). This is the standard form of the nonlinear lattice Boltzmann approximation (mean-field theory) for lattice gas automata.[17] The way in which this equation has been derived closely parallels the derivation of the nonlinear Boltzmann equation for hard spheres from the corresponding BBGKY hierarchy, where one replaces the pair distribution function $f^{(2)}$ in the first hierarchy equation by a product of single-particle distribution functions, or equivalently, one neglects the two-point correlation function.[22]

2.6. Generalized Boltzmann and Ring Equations

In *first* approximation (2.17) is inserted into (2.10), where the terms linear in G_{kl} are included in the kinetic equations, but higher-order terms are neglected. The nonlinear Boltzmann equation (2.19) is extended with a term linear in G, yielding the *generalized Boltzmann equation*,

$$f_i(\mathbf{r} + c_i, t) - f_i(\mathbf{r}, t) = \Omega_i^{(1,0)}(f(\mathbf{r}, t)) + \sum_{k<l} \Omega_{i,kl}^{(1,2)}(f(\mathbf{r}, t)) G_{kl}(\mathbf{r}, \mathbf{r}, t) \qquad (2.21)$$

The explicit form of the coefficients $\Omega_{i,kl}^{(1,2)} = \partial^2 \Omega_i^{(1,0)}/\partial f_k \, \partial f_l$ is given in (A.5). Equation (2.21) describes corrections to the mean-field equation (2.19) caused by the correlations G_{kl} that are built up by sequences of correlated ring collisions between the particles. These correlations can be calculated from the lowest-order approximation to the second hierarchy equation (see Appendix A). The result is the so-called *ring kinetic equation* for the pair correlation function,

$$G_{ij}(\mathbf{r} + \mathbf{c}_i, \mathbf{r}' + \mathbf{c}_j, t + 1)$$
$$= \sum_{k,l} \omega_{ij,kl}(f, f') G_{kl}(\mathbf{r}, \mathbf{r}', t) + \delta(\mathbf{r}, \mathbf{r}') B_{ij}(\mathbf{r}, t) \qquad (2.22)$$

where ω is the two-particle collision operator, given by

$$\omega_{ij,kl}(f,f') = \{\delta_{ik} + \Omega_{ik}^{(1,1)}(f(\mathbf{r},t))\}\{\delta_{jl} + \Omega_{jl}^{(1,1)}(f(\mathbf{r}',t))\} \tag{2.23}$$

The linearized Boltzmann operator, $\Omega_{ij}^{(1,1)}(f) = \partial\Omega_i^{(1,0)}/\partial f_j$, is given explicitly in (A.5). In particular, we have

$$\delta_{ik} + \Omega_{ik}^{(1,1)} = \sum_{s\sigma} \delta\sigma_i A_{s\sigma} F(s) \frac{\delta s_k}{g_k} \tag{2.24}$$

The on-node source term $B_{ij}(\mathbf{r},t)$ in (2.22) is a function of both $f_i(\mathbf{r},t)$ and $G_{ij}(\mathbf{r},\mathbf{r},t)$. It essentially contains the on-node postcollision correlations that are created by the collision step and originate from the last term on the right-hand side of (2.11). Furthermore, $B_{ij}(\mathbf{r},t)$ contains the term with $\delta(\mathbf{r},\mathbf{r}')$ on the second line of (2.11). The detailed expressions for these terms after cluster expansion are given in (A.11) and (A.16) of Appendix A.

The structure of the kinetic equations is essentially the same as for continuous fluids. Together, the generalized Boltzmann equation (2.21) and the ring equation (2.22) form a closed set of equations for the functions $f_i(\mathbf{r},t)$ and $G_{ij}(\mathbf{r},\mathbf{r}',t)$. These approximate kinetic equations still obey the global (standard and spurious) conservation laws. This is expressed by the *orthogonality* condition with respect to the collisional invariants a_i, i.e.,

$$\sum_{ij} a_i a_j B_{ij} = 0 \tag{2.25}$$

as follows from the definitions (A.11) and (A.16).

Equations (2.21) and (2.22) have been derived under the assumption that triplet- and higher-order correlations as well as products of pair correlations can be neglected. Only terms that are linear in the pair correlation occur. In principle more terms might be included in (2.21) and (2.22). The justification of our assumptions can only be given *a posteriori* by comparing the theoretical predictions with results from computer simulations. This will be done in the remainder of this paper, for the special case of equilibrium correlations.

3. EQUILIBRIUM CORRELATIONS

3.1. Ring Operator

The purpose of the present section is to obtain stationary solutions to the kinetic equations for the distribution and correlation functions under the assumption that a *spatially uniform equilibrium state* exists. The single-particle distribution function will be denoted by $f_i(\mathbf{r},\infty) = f_i$ and the pair correlations by $G_{ij}(\mathbf{r},\mathbf{r}',\infty) \equiv \mathcal{G}_{ij}(\mathbf{r}-\mathbf{r}')$.

For a spatially homogeneous equilibrium state, the kinetic equation (2.21) for f_i simplifies to

$$\Omega_i^{(1,0)}(f) + \sum_{k<l} \Omega_{i,kl}^{(1,2)}(f)\,\mathcal{G}_{kl}(\mathbf{0}) = 0 \tag{3.1}$$

The ring equation (2.22) for the pair correlations also simplifies. The source term B_{ij} in (2.22) is now independent of $\mathbf{r}$ and reduces to

$$B_{ij} = \mathcal{G}_{ij}^*(\mathbf{0}) - \sum_{k,l} \omega_{ij,kl}\mathcal{G}_{kl}(\mathbf{0}) \tag{3.2}$$

with

$$\mathcal{G}_{ij}^*(\mathbf{0}) = \Omega_{ij}^{(2,0)}(f) + \mathcal{G}_{ij}(\mathbf{0}) + \sum_{k<l} \Omega_{ij,kl}^{(2,2)}(f)\,\mathcal{G}_{k,l}(\mathbf{0}) \tag{3.3}$$

Here $\mathcal{G}_{ij}^*(\mathbf{0}) = G_{ij}^*(\mathbf{r}, \mathbf{r}, \infty)$ represents the on-node postcollision correlations in the stationary state. It is defined through (2.15) with δn_i in (2.14) replaced by the postcollision fluctuation $\delta n_i^* = n_i^* - f_i^*$. The expressions (3.2) and (3.3) can be derived from (A.11) and (A.16), where the contribution $B_{ij}^{(3)}$ in (A.11) vanishes in equilibrium, at least up to linear order in the pair correlations, due to (3.1), and the Ω coefficients are defined in (A.17).

In (3.3) the quantity

$$\Omega_{ij}^{(2,0)}(f) = \sum_{s\sigma} (\delta\sigma_i\delta\sigma_j - \delta s_i\delta s_j)\,A_{s\sigma}F(s) \tag{3.4}$$

accounts for the correlations created by a single collision from the completely factorized precollision state $F(s)$ in (2.18). It represents the dominant contribution to the on-node postcollision correlations. The next two terms represent the effect of existing precollision correlations. This second contribution is in general rather small.

The quantity $\Omega_{ij}^{(2,0)}$ was calculated earlier in ref. 16 and compared with on-node postcollision correlations measured in computer simulations. The data agreed within 10% for a triangular lattice gas violating semi-detailed balance. The present theory enables one to calculate the *precollision* correlations, which are built up by collective mechanisms, acting on large spatial and temporal scales. The ring operator (3.12) propagates the two-particle correlations to other nodes through uncorrelated single-particle motion (Boltzmann propagators) and recollects this information on a single node [represented by the $\mathbf{q}$ integration in (3.12)].

Further simplification is possible by introducing the Fourier transform,

$$\hat{\mathcal{G}}_{ij}(\mathbf{q}) = \sum_{\mathbf{r}} e^{-i\mathbf{q}\cdot\mathbf{r}}\mathcal{G}_{ij}(\mathbf{r}) \tag{3.5}$$

so that (2.22) becomes

$$\hat{\mathscr{G}}_{ij}(\mathbf{q}) - s_{ij}(\mathbf{q}) \sum_{k,l} \omega_{ij,kl} \hat{\mathscr{G}}_{ij}(\mathbf{q}) = s_{ij}(\mathbf{q}) B_{ij} \tag{3.6}$$

where the pair streaming or advection operator is defined as[6]

$$s_{ij}(\mathbf{q}) = \exp[-i\mathbf{q} \cdot (\mathbf{c}_i - \mathbf{c}_j)] \tag{3.7}$$

Using a matrix notation in which $\hat{\mathscr{G}}_{ij}(\mathbf{q})$ and B are represented as b^2-dimensional vectors with components $\hat{\mathscr{G}}_{ij}(\mathbf{q})$, etc., and ω, $s(\mathbf{q})$, and 1 as $b^2 \times b^2$ matrices with elements $\omega_{ij,kl}$, $s_{ij,kl}(\mathbf{q}) = s_{ij}(\mathbf{q})\delta_{ik}\delta_{jl}$, and $1_{ij,kl} = \delta_{ik}\delta_{jl}$, we can write (3.6) as

$$\{1 - s(\mathbf{q})\omega\} \hat{\mathscr{G}}(\mathbf{q}) = s(\mathbf{q})B \tag{3.8}$$

The formal solution to this equation is

$$\hat{\mathscr{G}}(\mathbf{q}) = \frac{1}{1 - s(\mathbf{q})\omega} s(\mathbf{q})B \tag{3.9}$$

However, there is a finite set of points $\mathbf{q}_n$, corresponding to global invariants, where the matrix $\{1 - s(\mathbf{q}_n)\omega\}$ has one or more zero eigenvalues, and therefore is not invertible. Careful analysis of this problem leads to a quantitative prediction of finite-size effects (see Appendix B). In what follows we will only consider the thermodynamic limit $(V \to \infty)$, where the contributions from the set $\{\mathbf{q}_n\}$ are of measure zero and can be ignored.

To solve the ring equation, we first note that according to (3.1) f_i couples only to the on-node correlations $\mathscr{G}(0)$. The method is therefore to first construct and solve coupled equations for f_i and $\mathscr{G}(0)$. Once these are known, B in (3.2) is completely determined. To achieve this we observe that the on-node correlations can be written in terms of Fourier components as

$$\mathscr{G}_{ij}(0) = \lim_{V \to \infty} \frac{1}{V} \sum_{\mathbf{q}} \hat{\mathscr{G}}_{ij}(\mathbf{q}) = v_0 \int \frac{d\mathbf{q}}{(2\pi)^d} \hat{\mathscr{G}}_{ij}(\mathbf{q}) \tag{3.10}$$

Here the $\mathbf{q}$ summation and integration run over the first Brillouin zone of the reciprocal lattice $\mathscr{L}^*$ and v_0 denotes the volume of a unit cell on the lattice (e.g., $v_0 = 1$ for the square and $v_0 = \frac{1}{2}\sqrt{3}$ for the triangular lattice).

---

[6] $s_{ij}(\mathbf{q})$ and $s(\mathbf{q})$ should not be confused with the occupation numbers $s_i(\mathbf{r}, t)$ or s_i used in Section 2.

Therefore integrating (3.9) over $\mathbf{q}$ gives the following *closed equation* for the on-node correlation matrix as a function of f:

$$\mathcal{G}_{ij}(\mathbf{0}) = \sum_{k,l} R_{ij,kl} B_{kl} \tag{3.11}$$

where B_{kl} in (3.2) contains terms linear in $\mathcal{G}(\mathbf{0})$. The ring operator R is defined as

$$R = v_0 \int \frac{d\mathbf{q}}{(2\pi)^d} \frac{1}{1 - s(\mathbf{q})\omega} s(\mathbf{q}) \tag{3.12}$$

With Eqs. (3.1), (3.2), and (3.11) we now have a closed description to determine both f_i and $\mathcal{G}_{ij}(\mathbf{0})$. Once the on-node correlations (and therefore also B_{kl}) have been solved from (3.11), then $\hat{\mathcal{G}}_{ij}(\mathbf{q})$ can be calculated using (3.9), and the full r-dependent correlation function $\mathcal{G}_{ij}(\mathbf{r})$ follows by inverse Fourier transformation of $\hat{\mathcal{G}}_{ij}(\mathbf{q})$,

$$\mathcal{G}_{ij}(\mathbf{r}) = v_0 \int \frac{d\mathbf{q}}{(2\pi)^d} e^{i\mathbf{q} \cdot \mathbf{r}} \hat{\mathcal{G}}_{ij}(\mathbf{q}) \tag{3.13}$$

In equilibrium the pre- and postcollision correlations are related by

$$\mathcal{G}_{ij}^*(\mathbf{r}) = \mathcal{G}_{ij}(\mathbf{r} + \mathbf{c}_i - \mathbf{c}_j) \tag{3.14}$$

as can be seen from the first equality in (2.11).

3.2. Numerical Evaluation

This section deals with the numerical evaluation of the theory of the previous subsection from an operational point of view. First, we describe how $\mathcal{G}_{ij}(\mathbf{0})$ can be calculated for given f_i. As the diagonal part of the on-node correlation function $\mathcal{G}_{ij}(\mathbf{0})$ is given by (2.16), it is convenient to introduce an equivalent description in terms of the *excess correlation* function $\mathcal{C}_{ij}$, defined by

$$\mathcal{C}_{ij} \equiv \mathcal{G}_{ij}(\mathbf{0}) - \mathcal{G}_{d,ij} \tag{3.15}$$

where $\mathcal{G}_{d,ij} = g_i \delta_{ij}$ with $g_i = f_i(1 - f_i)$. Combination of (3.2) and (3.11) yields a linear equation for the excess correlations,

$$\mathcal{C}_{ij} = \sum_{k,l} R_{ij,kl} \left\{ \Omega_{kl}^{(2,0)} + \sum_{m<n} \Omega_{kl,mn}^{(2,2)} \mathcal{C}_{mn} + \sum_{m \neq n} (1 - \omega)_{kl,mn} \mathcal{C}_{mn} \right\} \tag{3.16}$$

In deriving this equation we have used the relation $\mathscr{G}_d = R(1 - \omega)\,\mathscr{G}_d$, which is only valid in the thermodynamic limit (see Appendix B). Once R is known, we proceed to calculate $\mathscr{C}_{ij}$ for given f_i by splitting the ring equation (3.16) into an inhomogeneous part that depends only on f_i, and a part that is linear in $\mathscr{C}_{ij}$. Since $\mathscr{C}_{ij} = \mathscr{C}_{ji}$ is symmetric, the only independent elements of $\mathscr{C}_{ij}$ are those with $i < j$. These can conveniently be taken together as a vector in a $[\frac{1}{2}b(b-1)]$-dimensional space, spanned by the possible pairs (ij) with $i < j$. It is therefore appropriate to interpret (3.16) as a vector equation,

$$\mathscr{C} = K + M\mathscr{C} \tag{3.17}$$

where $\mathscr{C}$ and K are $[\frac{1}{2}b(b-1)]$-dimensional vectors with components labeled by the pair index (ij), and M is a matrix of equal dimensionality. It follows from (3.16) that

$$K_{(ij)} = \sum_{mn} R_{ij,mn}\Omega_{mn}^{(2,0)}$$

$$M_{(ij),(kl)} = \sum_{mn} R_{ij,mn}\left[\Omega_{mn,kl}^{(2,2)} + (1-\omega)_{mn,kl} + (1-\omega)_{mn,lk}\right] \tag{3.18}$$

Once K and M have been calculated for given f_i, the excess correlations are given by

$$\mathscr{C} = \frac{1}{1-M}K \tag{3.19}$$

provided that $\det(1 - M) \neq 0$.

The *simple ring* approximation to the precollision on-node correlation function is given by $\mathscr{C} = R\Omega^{(2,0)}$, which is equivalent to neglecting the term $M\mathscr{C}$ in (3.17). It captures the essential mechanism that is responsible for the pair correlations occurring in LGA violating detailed balance. However, there are corrections due to the fact that the on-node postcollision correlations are not generated from a factorized precollision state as described by $\Omega^{(2,0)}$, but from a correlated precollision state with pair correlations given by $\mathscr{C}_{ij}$. These corrections are represented by the term $M\mathscr{C}$ in (3.17), and give rise to the so-called *repeated ring* contribution to $\mathscr{C}_{ij}$. Numerical evaluation of $\mathscr{C}$, as given by (3.19), and comparison with the simple ring result $\mathscr{C} = R\Omega^{(2,0)}$ shows that the repeated ring corrections to the simple ring approximation are relatively small but significant (typically 5–10%).

In *single-speed* LGA, even if they violate detailed balance, lattice symmetries require that the single-particle distribution function is given by $f_i = \rho/b$. The presence of on-node correlations $\mathscr{C}_{ij}$ does not affect f_i, since $\Omega^{(1,2)}$ in (3.1) vanishes due to symmetry and $\mathscr{C}_{ij}$ can be calculated in a straightforward manner using (3.19).

In *multispeed* LGA (e.g., with additional rest particles) that violate semi-detailed balance, we have in general $f_i \neq \rho/b$. In the spirit of the basic assumption that all correlations are small, the *zeroth* approximation to f_i is given by the stationary solution f_i^0 of the nonlinear Boltzmann equation,

$$\Omega_i^{(1,0)}(f^0) = 0 \tag{3.20}$$

which is equivalent to setting the off-diagonal elements $\mathscr{C}_{ij}$ of $\mathscr{G}_{ij}(0)$ equal to zero in (3.1). We can calculate the off-diagonal pair correlations $\mathscr{C}_{ij}^0$ in first approximation by evaluating (3.19) with $f_i = f_i^0$; the diagonal correlations are given by $\mathscr{G}_{ii}^{(0)} = f_i^0(1 - f_i^0)$. In *first* approximation, the single-particle distribution function $f_i = f_i^0 + f_i^1$ can be obtained by evaluating the second term in (3.1) in first approximation, i.e., as $\Omega_{i,kl}^{(1,2)}(f^0) C_{kl}^0$, and solving (3.1) for f_i^1, which is a small correction to f_i^0.

We have found it efficient to use the following iterative scheme to obtain a self-consistent solution $\{f, \mathscr{C}\}$ to (3.1) and (3.19):

1. Find the stationary distribution f_i in Boltzmann approximation by solving (3.1) with $\mathscr{C}_{ij} \equiv 0$, i.e., $\Omega^{(1,0)}(f) = 0$. This can be done by iterating $f(n+1) = f(n) + \Omega^{(1,0)}(f(n))$, starting from $f_i = \rho/b$, until $\Omega^{(1,0)}(f) \simeq 0$.

2. Calculate $\mathscr{C}_{ij}$ from (3.19), using the values of f_i found in the previous step.

3. Find the stationary solution f_i of (3.1), using the values of $\mathscr{C}_{ij}$ found in the previous step. This can again be done by iterating $f(n+1) = f(n) + I(f(n))$, with $I = \Omega^{(2,0)} + \Omega^{(2,2)}\mathscr{C}$, until $I(f) \simeq 0$.

4. Repeat steps 2 and 3 until the scheme has converged.

We found that in most cases this scheme converged within a few iteration steps. The difference between the self-consistent value of f_i and the solution f_i^0 of the nonlinear Boltzmann equation (3.20) was in general rather small.

At this point we have completed our detailed description of how to obtain a spatially uniform stationary solution to the coupled time evolution equations for $f_i(t)$ and $\mathscr{G}_{ij}(\mathbf{r}, t)$. One can now ask whether this solution is unique. For single-speed models it clearly is, since for given $f_i = \rho/b$ the pair correlations $\mathscr{C}_{ij}(f)$ are uniquely determined by (3.19). However, in the case of multispeed models it cannot be excluded that there are other physically acceptable equilibrium solutions, as the Ω matrices are nonlinear functions of $f = \{f_i\}$. We can systematically search for stationary solutions by finding the zeros of $I_i(f) = \Omega_i^{(1,0)}(f) + \sum_{k<l} \Omega_{i,kl}^{(1,2)}(f) \mathscr{C}_{kl}(f)$ as a function of f. All solutions f of $I_i(f) = 0$ correspond to possible equilibrium states $\{f, C\}$.

3.3. Factorized Equilibrium States

For LGA satisfying the condition of semi-detailed balance with respect to the (universal) Gibbs equilibrium distribution, the matrix elements of $\Omega^{(2,0)}$ given by (3.4) vanish as a consequence of (2.4) together with the normalization (2.2). No correlations are created by the collision step in equilibrium, and both before and after collision the system is described by the completely factorized Gibbs distribution, parametrized by the average occupations f_i, which are given by a Fermi distribution that only depends on global invariants such as the average number of particles or (for fluid-type models) momentum per node. It follows from (3.19) that $\mathscr{C}_{ij} = 0$, and consequently $\mathscr{G}_{ij}(\mathbf{r} - \mathbf{r}') = \delta_{ij}\delta(\mathbf{r}, \mathbf{r}')f_i(1 - f_i)$.

As is less well known, even if condition (2.4) is violated there may still exist a completely factorized, but *nonuniversal* equilibrium state, with average occupations f_i that depend on the transition probabilities $A_{s\sigma}$. Examples can be found in ref. 16 and in Section 4.1 of this paper. The condition for the existence of such a nonuniversal equilibrium state is that a set of average occupations f_i can be found such that the corresponding factorized equilibrium distribution is invariant under the action of the collision step,[7]

$$F(\sigma) = \sum_s A_{s\sigma} F(s) \tag{3.21}$$

Equation (3.21) implies the relation $\Omega^{(2,0)}(f) = 0$, so that again $\mathscr{G}_{ij}(\mathbf{r} - \mathbf{r}') = \delta_{ij}\delta(\mathbf{r}, \mathbf{r}')f_i(1 - f_i)$, but now with nonuniversal f_i. It follows directly from the definition (2.20) of the nonlinear Boltzmann operator that (3.21) implies that $\Omega_i^{(1,0)} = 0$. In other words: if a factorized distribution F can be found that satisfies (3.21), it will be a *stationary* distribution. It should be stressed that in a closed (microcanonical) system, with finite L and fixed N, correlations $\mathscr{G}_{ij}(\mathbf{r})$ of $\mathcal{O}(1/V)$ exist that are independent of the distance $\mathbf{r}$, even in models *satisfying* semi-detailed balance. This can be seen as follows: for a semi-detailed balance model with V nodes and $N = bfV$ particles one has

$$\langle n_i(\mathbf{r}) \, n_j(\mathbf{r}') \rangle = f[(N - 1)/(bV - 1)] < f^2$$

[provided of course that $(\mathbf{r}, \mathbf{c}_i) \neq (\mathbf{r}', \mathbf{c}_j)$]. Consequently $\mathscr{G}_{ij}(\mathbf{r} - \mathbf{r}') = -f(1 - f)/(bV - 1) < 0$, independent of $\mathbf{r} - \mathbf{r}'$.

---

[7] This is an example of the more general semi-detailed balance or Stueckelberg condition, discussed in footnote 5.

4. APPLICATIONS

4.1. Interacting Random Walkers

The ring kinetic theory for approximate calculation of pair correlations presented in the previous sections assumes that all higher-order correlations are negligible. It is important to make a quantitative comparison with computer simulation results to establish the accuracy of the results obtained from a numerical evaluation of the theory.

We first consider a model of interacting random walkers on a line. An isolated random walker executes a persistent random walk while jumping to nearest neighbor sites or resting on the same site, i.e., it has three allowed velocity states, $c_i = \{ +1, 0, -1 \}$, with transition probabilities depending on the previous jump.

The interaction between the walkers is strictly local and essentially determined by the Fermi exclusion rule for different velocity channels, i.e., $n_i(r, t) = \{0, 1\}$. The maximum number of walkers on a site is therefore three. The transition probabilities from a two-particle in-state $s(r)$ to an out-state $\sigma(r)$ are defined in the right diagram of Fig. 1. There exists only a single three-particle state, which remains unchanged under interaction on account of the Fermi exclusion rule.

The above model can be described conveniently in terms of a one-dimensional diffusive LGA, where at every node one- and two-particle transitions occur with probabilities defined in Fig. 1. In principle there are 12 independent transition probabilities $A_{s\sigma} \neq 0$. This number is reduced to six if we require that the collision rules be invariant under reflection $(c_i \to -c_i)$. In the present model the total number of particles is conserved, but the total momentum is not, and there are no staggered or other spurious invariants. Consequently the model has only a single slow (diffusive) mode.

To illustrate some of the analytical results of the previous sections and to discuss the conditions of semi-detailed balance, it is of interest to write out the microdynamic equation (2.6) for the occupation numbers $n_+(r, t)$, $n_0(r, t)$, and $n_-(r, t)$, referring, respectively, to the velocity channels

Fig. 1. Transition probabilities in one-dimensional model of interacting random walkers with mutual exclusion in the same channel (r, c_i) with $c_i = \{ -1, 0, 1 \} = \{ \leftarrow, \bullet, \rightarrow \}$. The left and right diagrams show transitions between states with one and two particles, respectively.

$\{c_i = +1, 0, -1\}$. Let $n_i^*(r, t) = n_i(r + c_i, t + 1)$ be the postcollision occupation number; then

$$
\begin{aligned}
n_+^* &= n_+ + (\hat{\alpha}_r n_0 \bar{n}_+ - \hat{\beta} n_+ \bar{n}_0)\bar{n}_- + \hat{\gamma}(n_- \bar{n}_+ - n_+ \bar{n}_-)\bar{n}_0 \\
&\quad + (\hat{\beta}' n_0 \bar{n}_+ - \hat{\alpha}_l' n_+ \bar{n}_0)n_- + \hat{\gamma}'(n_- \bar{n}_+ - n_+ \bar{n}_-)n_0 \\
n_0^* &= n_0 + \hat{\beta}(n_+ \bar{n}_- + n_- \bar{n}_+)\bar{n}_0 - \hat{\alpha}_r n_0 \bar{n}_+ \bar{n}_- - \hat{\alpha}_l n_0 \bar{n}_+ \bar{n}_- \\
&\quad + \hat{\alpha}_r' n_+ n_- \bar{n}_0 + \hat{\alpha}_l' n_+ n_- \bar{n}_0 - \hat{\beta}'(n_+ \bar{n}_- + n_- \bar{n}_+)n_0
\end{aligned}
\tag{4.1}
$$

and a similar relation for n_-^*. The set $\hat{A}_{s\sigma} = \{\hat{\alpha}_r, \hat{\alpha}_l, \hat{\alpha}_r', \hat{\alpha}_l', \hat{\beta}, \hat{\beta}', \hat{\gamma}, \hat{\gamma}'\}$ represents the Boolean realizations $\hat{A}_{s\sigma}(r, t)$ discussed below (2.6), with expectation values $A_{s\sigma} = \{\alpha_r, \alpha_l, \alpha_r', \alpha_l', \beta, \beta', \gamma, \gamma'\}$. The subscripts $\{r, l\}$ refer to transitions to states with a particle moving to the right and to the left, respectively. As mentioned earlier, reflection symmetry imposes that $\alpha_r = \alpha_l = \alpha$ and $\alpha_r' = \alpha_l' = \alpha'$. The model can be made self-dual (invariant under exchanging particles and holes) by choosing $\alpha = \alpha'$, $\beta = \beta'$, and $\gamma = \gamma'$. However, we keep the distinction between α and α', etc., in order to discuss the distinction between detailed balance with respect to the universal Gibbs distribution and to a nonuniversal stationary distribution, respectively.

Consider first the mean-field or Boltzmann approximation. According to (2.19) the nonlinear Boltzmann equation for $f_i(r, t)$ is obtained from (4.1) by replacing $n_i(r, t)$ by $f_i(r, t)$ and the Boolean variables $\hat{\alpha}$, etc., by their expectation values α, etc. In equilibrium the distribution function f_i is characterized by the average occupation numbers f_0 and $f_+ = f_-$ with average density $\rho = f_0 + 2f_+$.

For this model the condition for semi-detailed balance (2.4) and detailed balance (2.5) with respect to the Gibbs distribution are equivalent, and given by

$$
\alpha = \beta, \qquad \alpha' = \beta'
\tag{4.2}
$$

We recall that the stationary distribution is a Gibbs distribution if the phase space density depends only on the conserved quantities, so that $f_i = \frac{1}{3}\rho$, independent of the velocity c_i.

As explained in Section 3.4, it is also possible to have a completely factorized but nonuniversal equilibrium state, where the average occupations f_i depend on the transition probabilities $A_{s\sigma}$. Writing out (3.20) and using $f_- = f_+$, we obtain

$$
\begin{aligned}
\alpha f_0 (1 - f_+)^2 &= \beta (1 - f_0) f_+ (1 - f_+) \\
\beta' f_0 f_+ (1 - f_+) &= \alpha' (1 - f_0) f_+^2
\end{aligned}
\tag{4.3}
$$

These two equations can only be satisfied simultaneously if

$$\alpha\alpha' = \beta\beta' \tag{4.4}$$

In that case f_0 and $f_+ = f_-$ are related by $z_0/z_+ = \beta/\alpha = \alpha'/\beta'$, with $z_i = f_i/(1 - f_i)$. At a given density ρ we can then use $\rho = f_0 + 2f_+$ to obtain a cubic equation for f_0.

If condition (4.4) is violated, there exist correlations between the occupation numbers of the different channels $(\mathbf{r}, \mathbf{c}_i)$, even in the equilibrium state. The critical quantity that determines whether equilibrium correlations are nonvanishing is $\Omega_{ij}^{(2,0)}(f)$, defined in (3.4). For the present model we have

$$\begin{aligned}
\Omega_{+-}^{(2,0)}(f) &= 2(\beta'f_0\bar{f}_+ - \alpha'f_+\bar{f}_0)f_+ \\
\Omega_{+0}^{(2,0)}(f) &= \Omega_{-0}^{(2,0)}(f) = (\alpha'f_+\bar{f}_0 - \beta'f_0\bar{f}_+)f_+
\end{aligned} \tag{4.5}$$

If the transition probabilities satisfy detailed balance with respect to the Gibbs distribution (4.2) or a nonuniversal factorized distribution (4.4), then $\Omega_{ij}^{(2,0)}(f) = 0$, and all on- and off-node correlations vanish.

The method developed in the present paper enables one to calculate the on-node quantities $\{f_i, \mathscr{C}_{ij}\}$ numerically. The off-node correlations $\mathscr{G}_{ij}(r)$ can then be constructed with the help of (3.9) and (3.13). The postcollision correlations $\mathscr{G}_{ij}^*(r)$ are then given by (3.14). Note that for the three-bit model $\mathscr{G}_{-+}^* = \mathscr{G}_{ij}(2)$ and $\mathscr{G}_{0+}^* = \mathscr{G}_{0+}(1)$, where $r = 1$ and $r = 2$ denote the nearest- and next-nearest-neighbor sites. The relative importance of the pre- and postcollision pair correlations $\mathscr{G}_{ij}$ and $\mathscr{G}_{ij}^*$ is best measured when they are normalized by the single-channel fluctuations $g_i = \langle(\delta n_i)^2\rangle$, yielding the covariances[8]

$$\text{Cov}(i,j) = \frac{\mathscr{G}_{ij}}{(g_i g_j)^{1/2}}, \qquad \text{Cov}^*(i,j) = \frac{\mathscr{G}_{ij}^*}{(g_i g_j)^{1/2}} \tag{4.6}$$

Using the method of Section 3.2, we have calculated these quantities for different choices of reduced density $f = \rho/3$, lattice size L, and transition probabilities α, β, γ (for self-dual models only), and compared the theoretical results with computer simulations.

In Table I a comparison is made between the different levels of sophistication at which the ring kinetic theory can be evaluated numerically: (i) either using the solution f_i^0 of the nonlinear Boltzmann equation (3.20), or using the self-consistent solution f_i obtained with the iterative scheme of

---

[8] In ref. 16 the correlations $\mathscr{G}_{ij}$ were normalized by $f_i f_j$ rather than by $(g_i g_j)^{1/2}$ as is done in this paper. Only with the latter choice is the self-dual symmetry preserved.
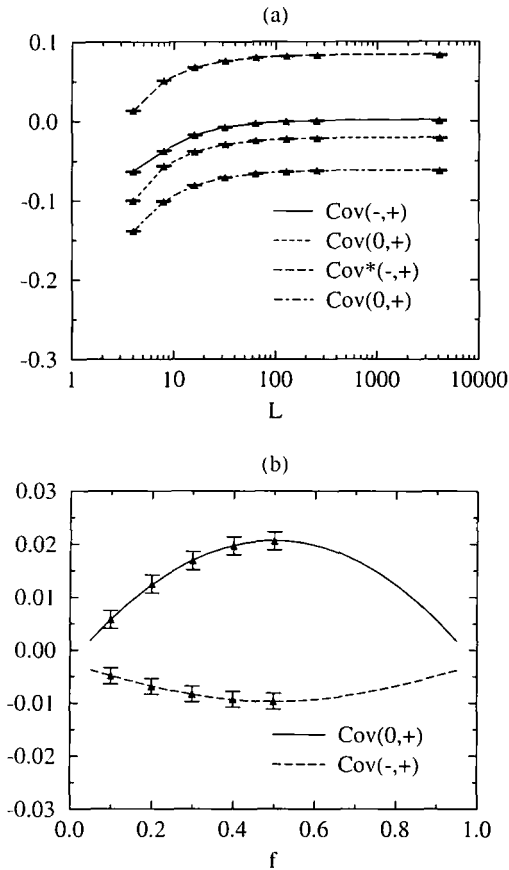
Fig. 2. Comparison of computer simulation results (symbols with error bars) and self-consistent ring kinetic theory (lines) for the model of Fig. 1. On-node correlations, indicated as covariances, are plotted (a) versus system size L, for transition probabilities $\alpha = 0.4$, $\beta = 0.5$, and $\gamma = 0$ and density $f = 0.5$; (b) versus density f, for $L = 128$, $\alpha = 0.5$, $\beta = 0.4$, and $\gamma = 0$; (c) versus transition probability α, for $f = 0.5$, $L = 128$, $\beta = 0.33$, and $\gamma = 0.5$. Note the vanishing of correlations when $\alpha = \beta$ (detailed balance).

Section 3.2, and (ii) either using the simple ring approximation or including repeated ring contributions as well. For reference we have included the values of the postcollision correlations calculated in ref. 16. They are equivalent to the simplest approximation (i.e., Boltzmann f^0, simple ring). The comparison with simulation data shows that the most sophisticated method (i.e., self-consistent, repeated ring) is indeed the most accurate. For an accurate predicion of pair correlations it appears to be more
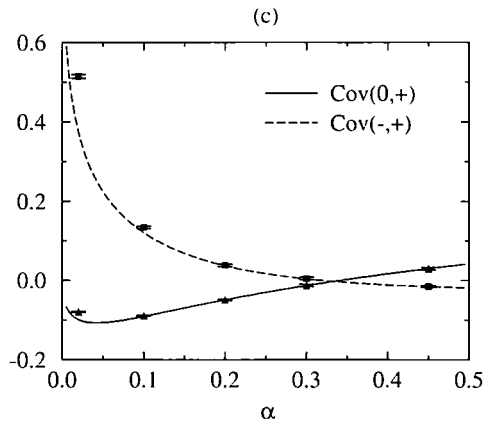
(c)

Fig. 2. *(Continued)*

important to include repeated ring effects than to take the deviation of the self-consistent f_i from f_i^0 into account.

In Figure 2a values of the pre- and postcollision on-node correlations, obtained both from the numerical evaluation of the theory (lines) and from computer simulations (symbols with error bars), are plotted as a function of the system size L. For small values of L there are strong finite-size effects, which are predicted quantitatively by the theory of Appendix B. Figure 2b shows the typical dependence on the reduced density $f = \rho/3$; the correlation functions are symmetric around $f = 0.5$, as a consequence of the imposed self-duality. In Fig. 2c the on-node correlations are plotted as a function of α, for fixed $\beta = 0.33$ and $\gamma = 0.5$ at reduced density $f = 0.5$ and

Table I. Comparison of Methods for Calculating Static Pair Correlations in Equilibrium for the One-Dimensional 3-Bit Model Defined in Fig. 1[a]

Method	f_0	$f_{\pm}$	Cov_{0+}	Cov_{-+}	Cov_{0+}^{*}	Cov_{-+}^{*}
Theory of ref. 16	0.2578	0.3211	0	0	+0.1324	−0.2480
Boltzmann f_i^0						
Simple ring	0.2578	0.3211	+0.0738	−0.0219	+0.1324	−0.2480
Repeated ring	0.2578	0.3211	+0.0671	−0.0200	+0.1597	−0.1949
Self-consistent f_i						
Simple ring	0.2620	0.3190	+0.0727	−0.0225	+0.1323	−0.2483
Repeated ring	0.2617	0.3192	+0.0665	−0.0206	+0.1593	−0.1956
Simulation	0.2623	0.3200	+0.0654	−0.0218	+0.1584	−0.1970

[a] All data refer to collision parameters $\alpha = 0.5$, $\beta = 0.2$, and $\gamma = 0.5$, reduced density $f = 0.3$, and system size $L = 256$.

$L = 128$. Note that when $\alpha = \beta$ all correlations vanish: the model satisfies (semi-) detailed balance.

Since there is no fundamental difference between on- and off-node correlations, and since in addition the L dependence of the correlations is well predicted by the theory, we expect that our theory is capable of giving good predictions for off-node correlations $\mathscr{G}_{ij}(r)$ as well. This expectation is justified by Fig. 3, where for fixed $f = 0.5$, $L = 128$, and two different sets of transition probabilities, the density–density correlation function $\mathscr{G}(r) = \sum_{ij} \mathscr{G}_{ij}(r)$ is plotted versus r. Note that $\mathscr{G}(r)$ is negative at large r, due to the finite-size effects referred to in Section 3.3.
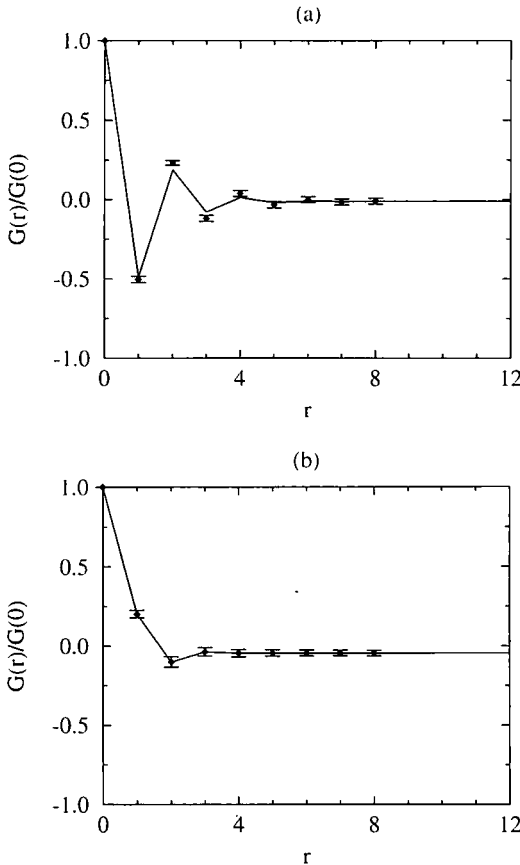

Fig. 3. Off-node density–density correlation function $G(r) = \sum_{ij} G_{ij}(r)$ plotted versus r, for $f = 0.5$, $L = 256$, and two different choices of transition probabilities: (a) $\alpha = 0.1$, $\beta = 0.5$, $\gamma = 0.5$ and (b) $\alpha = 0.5$, $\beta = 0.1$, $\gamma = 0.5$. Simulations (symbols with error bars) compared with theoretical values (solid line connecting points $r = 0, 1, 2,...$).

To study how the system approaches equilibrium in a spatially uniform nonequilibrium state we have solved the time-dependent evolution equations (2.21) and (2.22). Figure 4a shows how the typical relaxation time increases with L. Note that for $t < L/2$ the evolution is independent of L, which can be explained by the observation that the shortest time in which two opposite moving particles can meet through the periodic boundary conditions is $t = L/2$; at earlier times there is no distinction between a finite and an infinite system. When $L = 1024$ the system still has not reached equilibrium after 1000 time steps.
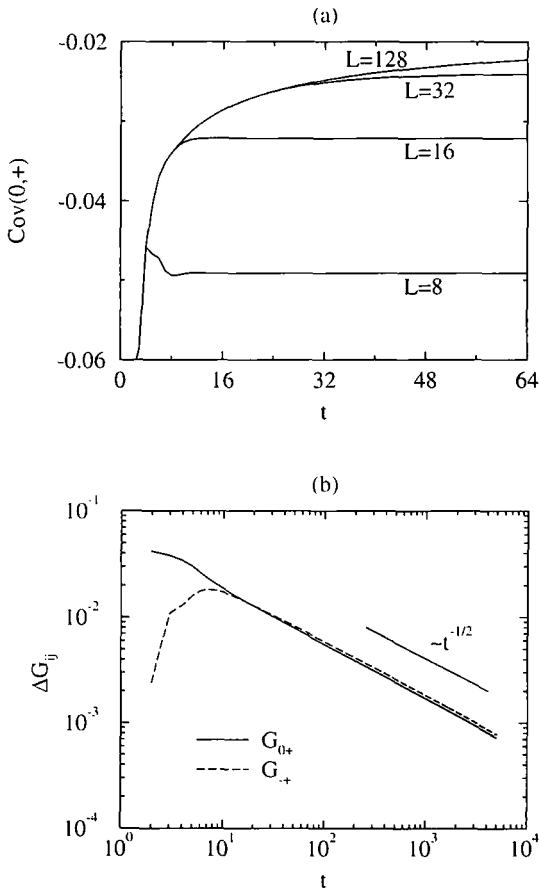

Fig. 4. Time dependence of on-node correlations, calculated using ring kinetic equation (no simulations), with $f = 0.5$ and the transition probabilities of Fig. 2a: (a) short-time behavior for various L; (b) algebraic long-time behavior, $\Delta G_{ij} \sim t^{-1/2}$, for $L = 4096$.

For long times the approach to equilibrium is algebraic,

$$\mathscr{G}_{ij}(\mathbf{0}, t) - \mathscr{G}_{ij}(\mathbf{0}, \infty) \equiv \Delta\mathscr{G}_{ij}(t) \sim t^{-\alpha} \tag{4.7}$$

with an exponent α that depends on the dimensionality and the type of collisional invariants of the mode. Figure 4b shows that $\alpha = 1/2$ in the case of the interacting random walkers, where the number of particles is the only collisional invariant. In a separate publication we will analyze the exponents and amplitudes for these algebraic tails in detail.

4.2. Two-Dimensional Fluid-Type Model

As a second application we consider a stochastic modification of the model introduced by Frisch *et al.*[17] that violates detailed balance. In this two-dimensional model, which was first introduced in ref. 16, particles move on a triangular lattice. Each node may contain a rest particle and up to six moving particles. The transition matrix $A_{s\sigma}$ introduced in Section 2 may be *asymmetric* for transitions between a state with and one without a rest particle. The general model contains 20 independent matrix elements $A_{s\sigma}$, as illustrated in Fig. 5.

In ref. 16 we obtained $\mathscr{C}_{ij} = 0$ and $\mathscr{C}_{ij}^* = \Omega_{ij}^{(2,0)}$ as simple estimates for the pre- and postcollision correlations, respectively, but a theory for non-vanishing precollision correlations in equilibrium was entirely lacking. The present paper provides the missing theory. Here we present the application of the present theory to this model, for a particular choice of collision
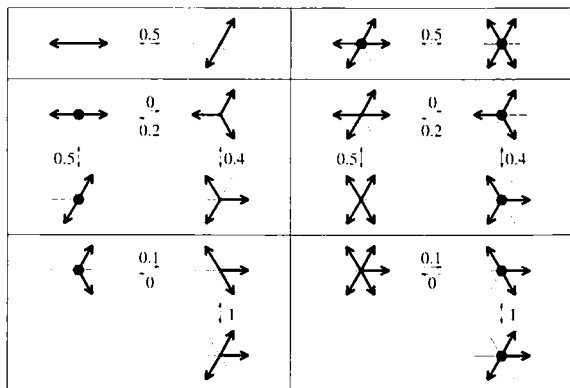

Fig. 5. Definition of the two-dimensional fluid-type LGA discussed in Section 4.2. Some transitions are not shown, for clarity; they can easily be reconstructed by noting that the collision rules must be invariant under rotations, e.g., to the transition in the upper left corner the clockwise rotation of the left state must be added.
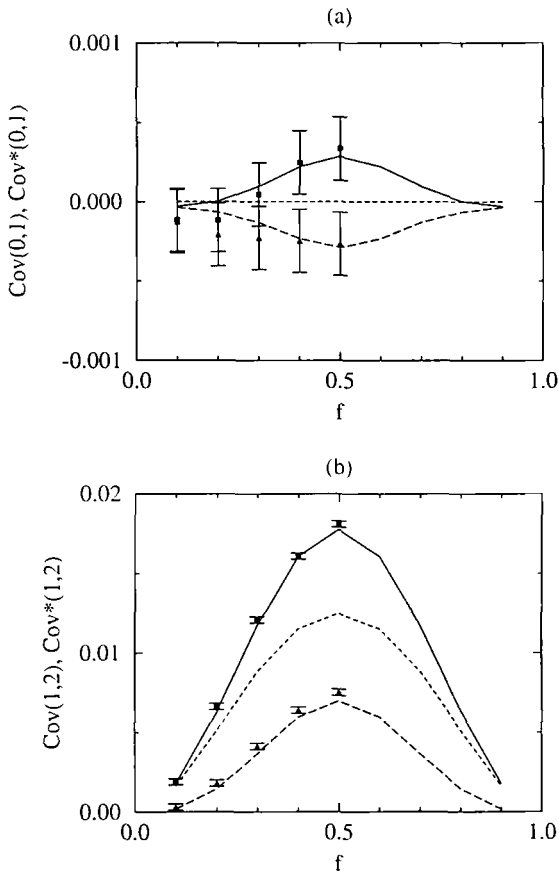
Fig. 6. Test of ring kinetic theory for the model of ref. 16, using the transition probabilities of set #13. Simulation data for $L = 64$ (symbols with error bars), compared with old theory for on-node postcollision correlations of ref. 16 (dotted line), and with new ring kinetic theory for the on-node precollision (dashed line) and postcollision (solid line) correlations.

parameters shown in Fig. 5 (in ref. 16 this choice of collision parameters was referred to as "set #13"). For this parameter set the correlations reach a stationary value after a few hundred time steps.

The theoretical predictions for the correlation functions have been compared with computer simulations of high statistical accuracy, taking several hours of CPU time on a Sparc 10 workstation for each data point. In basic equilibrium, where the total momentum **P** vanishes, the only independent elements of $\mathscr{C}_{ij}$ are $(ij) = \{(01), (12), (13), (14)\}$; all other elements are related to these four by lattice symmetries. As the model defined in
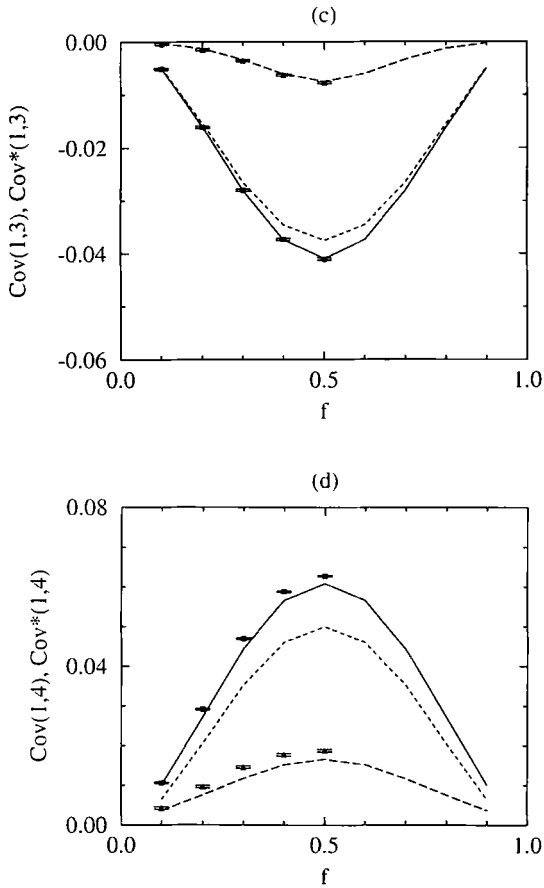
(c)

(d)

Fig. 6.   *(Continued)*

Fig. 5 is self-dual, the correlations are symmetric around $f = \rho/7 = 0.5$. Figure 6 shows the simulation results (symbols) compared with the ring kinetic theory for precollision (solid line) and postcollision (dashed line) correlations. From this figure it is clear that the self-consistent ring kinetic theory agrees very well with the simulations. For comparison we also plotted the old theory for the postcollision correlations of ref. 16 (dotted line).

5. CONCLUSION

   In the present paper a theory has been presented to calculate distri-bution and correlation functions in the stationary state of lattice gas

automata (LGA) violating semi-detailed balance. The theory of standard LGA satisfying semi-detailed balance is recovered as a special case.

The ring operator R defined in (3.12) that is used to calculate equilibrium correlations is identical to the one introduced for calculating long-time tails of time correlation functions[4,9] and corrections to Boltzmann transport coefficients[5] in LGA satisfying semi-detailed balance. One can show that linearization of the theory in this paper around the Gibbsian equilibrium state for standard LGA, together with neglect of repeated ring contributions, reproduces the results of refs. 4, 5, and 9.

In Section 3 we discussed an iterative scheme for finding a self-consistent solution $\{f_i, \mathscr{C}_{ij}\}$, starting from the solution of the nonlinear Boltzmann equation (where $\mathscr{C}_{ij} \equiv 0$). Even though the ring equation uniquely determines $\mathscr{C}_{ij}$ for given f_i, it is highly nonlinear in f_i; therefore it can in principle not be excluded that more than one spatially uniform stationary state exists.

To test our theory we have applied it to two different LGA, both violating semi-detailed balance. The theory presented in this paper shows excellent agreement with the results of computer simulations, both for a one-dimensional diffusive LGA and for a two-dimensional fluid-type LGA. Moreover, the theory accounts very well for finite-size effects.

As $t \to \infty$ a stationary state is approached. Due to the existence of local conservation laws the approach to this stationary state is algebraic, $\sim t^{-\alpha}$, with an exponent α that depends on the dimensionality and on the type of conservation laws. There is an intimate connection between the algebraic tails in the approach toward the correlated equilibrium state mentioned here and the well-known long-time tails in the decay of the velocity autocorrelation function.[23,24]

It would be of interest to apply the method of this paper to cellular automaton models for chemical reactions[25] or traffic flow,[26] where static correlations may play an important role.

It is appropriate to give some comments on the validity of the generalized Boltzmann equation and ring equation derived in this paper. In real fluids the validity of the cluster expansion is restricted to dilute systems, where the density acts as a small expansion parameter. Here there is no such small parameter and the approximations are less controlled. To understand the great success of the present mean-field theory, it is useful to compare the lattice Boltzmann equation with the revised Enskog theory for hard-sphere fluids.[27] Both theories describe nonequilibrium phenomena reasonably well over the whole density range. The reason for this seems to be that the collision term for the revised Enskog theory (resp. lattice Boltzmann equation) contains the *static correlations* imposed by the hard-core exclusion (resp. Fermi exclusion), which are the origin for the strong

density dependence of the theory. The present ring kinetic theory equation also contains—through the expansion coefficients Ω—all static correlations between colliding particles imposed by the Fermi exclusion. This might explain the success in predicting the density dependence of the correlation functions.

APPENDIX A. CLUSTER EXPANSION

In this appendix the cluster expansion of the hierarchy equations is derived. We start by substituting $n_i = f_i + \delta n_i$, resp. $s_i = f_i + \delta s_i$ into (2.12) and obtain the identity

$$(f_i + \delta n_j)^{s_j} (1 - f_j - \delta n_j)^{1 - s_j} = f_j^{s_j} (1 - f_j)^{1 - s_j} \left[1 + \frac{\delta s_j \, \delta n_j}{g_j} \right] \quad (A.1)$$

with $g_j = f_j(1 - f_j)$. With the help of this identity we can expand the single-node distribution $p(s, \mathbf{r}, t)$ defined in (2.9),

$$p(s, \mathbf{r}, t) = F(s, \mathbf{r}, t) \left\{ 1 + \sum_{k < l} \frac{\delta s_k(\mathbf{r}, t) \, \delta s_l(\mathbf{r}, t)}{g_k(\mathbf{r}, t) \, g_l(\mathbf{r}, t)} G_{kl}(\mathbf{r}, t) + \cdots \right\} \quad (A.2)$$

where $F(s, \mathbf{r}, t)$ is the factorized single-node distribution function (2.18). Similarly we can expand the two-node distribution function in (2.9) as

$$p^{(2)}(s, \mathbf{r}, s', \mathbf{r}', t) - p(s, \mathbf{r}, t) \, p(s', \mathbf{r}', t)$$

$$= F(s, \mathbf{r}, t) \, F(s', \mathbf{r}', t) \sum_{k, l} \frac{\delta s_k(\mathbf{r})}{g_k(\mathbf{r}, t)} \frac{\delta s_l(\mathbf{r}')}{g_l(\mathbf{r}', t)} G_{kl}(\mathbf{r}, \mathbf{r}', t) + \cdots \quad (A.3)$$

Insertion of (A.2) in (2.10) yields

$$f_i(\mathbf{r} + \mathbf{c}_i, t + 1) - f_i(\mathbf{r}, t) \simeq \Omega_i^{(1,0)}(f) + \sum_{k < l} \Omega_{i, kl}^{(1,2)}(f) \, G_{kl}(\mathbf{r}, \mathbf{r}, t) \quad (A.4)$$

The expansion coefficients Ω are given by

$$\Omega_i^{(1,0)}(f) = \sum_{s\sigma} (\sigma_i - s_i) \, A_{s\sigma} F(s)$$

$$\Omega_{ij}^{(1,1)}(f) = \sum_{s\sigma} (\sigma_i - s_i) \, A_{s\sigma} F(s) \frac{\delta s_j}{g_j} \quad (A.5)$$

$$\Omega_{i, kl}^{(1,2)}(f) = \sum_{s\sigma} (\sigma_i - s_i) \, A_{s\sigma} F(s) \frac{\delta s_k \, \delta s_l}{g_k \, g_l}$$

where $k < l$ and $F(s) = F(s, \mathbf{r}, t)$. Moreover, $\delta\sigma_i = \sigma_i - f_i$. Note that the Ω's depend on $\mathbf{r}$ and t through $f(\mathbf{r}, t)$. They are generated by the Taylor expansion of the nonlinear Boltzmann collision operator, $\Omega_i^{(1,0)}(f + \delta f)$, in powers of δf,

$$\Omega_i^{(1,0)}(f + \delta f) = \Omega_i^{(1,0)}(f) + \sum_j \Omega_{ij}^{(1,1)}(f)\, \delta f_j$$

$$+ \tfrac{1}{2} \sum_{k,l} \Omega_{ij}^{(1,2)}(f)\, \delta f_k\, \delta f_l + \cdots \qquad (A.6)$$

To derive an equation of motion for the precollision correlation function $G_{ij}(\mathbf{r}, \mathbf{r}', t)$ we start from its definition (2.15), written as

$$G_{ij}(\mathbf{r} + \mathbf{c}_i, \mathbf{r}' + \mathbf{c}_j, t + 1)$$
$$= f_{ij}^{(2)}(\mathbf{r} + \mathbf{c}_i, \mathbf{r}' + \mathbf{c}_j, t + 1)$$
$$- f_i(\mathbf{r} + \mathbf{c}_i, t + 1) f_j(\mathbf{r}' + \mathbf{c}_j, t + 1) \qquad (A.7)$$

and use (2.10) together with (2.11) to obtain

$$G_{ij}(\mathbf{r} + \mathbf{c}_i, \mathbf{r}' + \mathbf{c}_j, t + 1)$$
$$= [1 - \delta(\mathbf{r}, \mathbf{r}')] \sum_{s\sigma} \sum_{s'\sigma'} \sigma_i \sigma_j' A_{s\sigma} A_{s'\sigma'}$$
$$\times \{ p^{(2)}(s, \mathbf{r}, s', \mathbf{r}', t) - p(s, \mathbf{r}, t)\, p(s', \mathbf{r}', t) \}$$
$$+ \delta(\mathbf{r}, \mathbf{r}') \left\{ \sum_{s\sigma} \sigma_i \sigma_j A_{s\sigma}\, p(s, \mathbf{r}, t) \right.$$
$$\left. - \sum_{s\sigma} \sum_{s'\sigma'} \sigma_i \sigma_j' A_{s\sigma} A_{s'\sigma'}\, p(s, \mathbf{r}, t)\, p(s', \mathbf{r}, t) \right\} \qquad (A.8)$$

Next one verifies that σ_l $(l = i, j)$ and σ_j' can be replaced by $\delta\sigma_l = \sigma_l - f_l(\mathbf{r}, t)$ and $\delta\sigma_j' = \sigma_j' - f_j(\mathbf{r}', t)$ respectively. This leads to the form

$$G_{ij}(\mathbf{r} + \mathbf{c}_i, \mathbf{r}' + \mathbf{c}_j, t + 1) = K_{ij}(\mathbf{r}, \mathbf{r}', t) + \delta(\mathbf{r}, \mathbf{r}')\, B_{ij}(\mathbf{r}, t) \qquad (A.9)$$

with

$$K_{ij}(\mathbf{r}, \mathbf{r}', t) = \sum_{s\sigma} \sum_{s'\sigma'} \delta\sigma_i\, \delta\sigma_j'\, A_{s\sigma} A_{s'\sigma'}$$
$$\times \{ p^{(2)}(s, \mathbf{r}, s', \mathbf{r}', t) - p(s, \mathbf{r}, t)\, p(s', \mathbf{r}', t) \} \qquad (A.10)$$

and the on-node source term,

$$B_{ij}(\mathbf{r}, t) = B_{ij}^{(1)}(\mathbf{r}, t) + B_{ij}^{(2)}(\mathbf{r}, t) + B_{ij}^{(3)}(\mathbf{r}, t) \qquad (A.11)$$

consisting of three parts,

$$B_{ij}^{(1)}(\mathbf{r}, t) = \sum_{s\sigma} \delta\sigma_i \, \delta\sigma_j \, A_{s\sigma} \, p(s, \mathbf{r}, t)$$

$$= G_{ij}(\mathbf{r}, \mathbf{r}, t) + \sum_{s\sigma} (\delta\sigma_i \, \delta\sigma_j - \delta s_i \, \delta s_j) \, A_{s\sigma} \, p(s)$$

$$B_{ij}^{(2)}(\mathbf{r}, t) = -\sum_{s\sigma} \sum_{s'\sigma'} \delta\sigma_i \, \delta\sigma_j' \, A_{s\sigma} A_{s'\sigma'}$$

$$\times \left[p^{(2)}(s, \mathbf{r}, s', \mathbf{r}, t) - p(s, \mathbf{r}, t) \, p(s', \mathbf{r}, t) \right]$$

$$B_{ij}^{(3)}(\mathbf{r}, t) = -\sum_{s\sigma} \sum_{s'\sigma'} \delta\sigma_i \, \delta\sigma_j' \, A_{s\sigma} A_{s'\sigma'} \, p(s, \mathbf{r}, t) \, p(s', \mathbf{r}, t) \qquad (\text{A.12})$$

Inspection of (A.8) and (A.12) shows that the last two terms in (A.8) represent the postcollision on-node correlations,

$$G_{ij}^*(\mathbf{r}, \mathbf{r}, t) = B^{(1)}(\mathbf{r}, t) + B^{(3)}(\mathbf{r}, t) \qquad (\text{A.13})$$

whereas $B^{(2)}(\mathbf{r}, t)$ represents a correction coming from the off-node correlations. The cluster expansion of K_{ij} follows by inserting (A.3) into (A.10) with the result

$$K_{ij}(\mathbf{r}, \mathbf{r}', t) \simeq \sum_{k, l} \omega_{ij, kl}(f, f') \, G_{kl}(\mathbf{r}, \mathbf{r}', t) \qquad (\text{A.14})$$

where the pair collision operator is defined as

$$\omega_{ij, kl}(f, f') = \sum_{s\sigma s'\sigma'} \delta\sigma_i \, \delta\sigma_j' \, A_{s\sigma} A_{s'\sigma'} F(s) \, F(s') \frac{\delta s_k \, \delta s_l'}{g_k \, g_l'}$$

$$= \{ \delta_{ik} + \Omega_{ik}^{(1, 1)}(f(\mathbf{r}, t)) \} \{ \delta_{jl} + \Omega_{jl}^{(1, 1)}(f(\mathbf{r}', t)) \} \qquad (\text{A.15})$$

In the last line of (A.15) relation (A.5) has been used. For the three terms in $B_{ij}(\mathbf{r}, t)$ we obtain

$$B_{ij}^{(1)}(\mathbf{r}, t) \simeq G_{ij}(\mathbf{r}, \mathbf{r}, t) + \Omega_{ij}^{(2, 0)}(f) + \sum_{k < l} \Omega_{ij, kl}^{(2, 2)}(f) \, G_{kl}(\mathbf{r}, \mathbf{r}, t)$$

$$B_{ij}^{(2)}(\mathbf{r}, t) \simeq -\sum_{k, l} \omega_{ij, kl}(f, f) \, G_{kl}(\mathbf{r}, \mathbf{r}, t)$$

$$\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad (\text{A.16})$$

$$B_{ij}^{(3)}(\mathbf{r}, t) \simeq -\Omega_i^{(1, 0)}(f) \, \Omega_j^{(1, 0)}(f)$$

$$- \sum_{k < l} \{ \Omega_i^{(1, 0)}(f) \, \Omega_{j, kl}^{(1, 2)}(f) + \Omega_j^{(1, 0)}(f) \, \Omega_{i, kl}^{(1, 2)}(f) \} \, G_{kl}(\mathbf{r}, \mathbf{r}, t)$$

We have introduced two more expansion coefficients, where $k < l$,

$$\Omega_{ij}^{(2, 0)}(f) = \sum_{s\sigma} (\delta\sigma_i \, \delta\sigma_j - \delta s_i \, \delta s_j) \, A_{s\sigma} F(s)$$

$$\Omega_{ij, kl}^{(2, 2)}(f) = \sum_{s\sigma} (\delta\sigma_i \, \delta\sigma_j - \delta s_i \, \delta s_j) \, A_{s\sigma} F(s) \frac{\delta s_k \, \delta s_l}{g_k \, g_l} \qquad \text{(A.17)}$$

APPENDIX B. FINITE-SIZE EFFECTS

Consider the formal solution of the ring equation (3.8),

$$\hat{\mathscr{G}}(\mathbf{q}) = \frac{1}{1 - s(\mathbf{q})\omega} \, s(\mathbf{q}) \, B \qquad \text{(B.1)}$$

Using a biorthogonal set of right and left eigenvectors, $\tilde{\chi}_\alpha(\mathbf{q})$ and $\chi_\alpha(\mathbf{q})$, satisfying

$$\langle \chi_\alpha(\mathbf{q}) \mid \tilde{\chi}_\beta(\mathbf{q}) \rangle \equiv \sum_{ij} \langle \chi_{\alpha, ij}(\mathbf{q}) \mid \tilde{\chi}_{\beta, ij}(\mathbf{q}) \rangle = \delta_{\alpha\beta}$$

and eigenvalues $\lambda_\alpha(\mathbf{q})$ of the two-particle propagator $s(\mathbf{q})\omega$, we can write $(1 - s(\mathbf{q})\omega)^{-1}$ as a spectral decomposition,

$$\left[\frac{1}{1 - s(\mathbf{q})\omega} \right]_{ij, kl} = \sum_\alpha \tilde{\chi}_{\alpha, ij}(\mathbf{q}) \frac{1}{1 - \lambda_\alpha(\mathbf{q})} \chi_{\alpha, kl}(\mathbf{q}) \qquad \text{(B.2)}$$

provided that $\lambda_\alpha(\mathbf{q}) \neq 1$. There is, however, a complication if, for one or more reciprocal lattice vectors $\mathbf{q}_n$ belonging to a set $\{\mathbf{q}_n\}$, there are eigenmodes α for which $\lambda_\alpha(\mathbf{q}_n) = 1$. The operator $(1 - s(\mathbf{q}_n)\omega)$ then has a null space $\mathcal{N}_n$, spanned by the eigenvectors for which $\lambda_\alpha(\mathbf{q}_n) = 1$. The set of points $\{\mathbf{q}_n\}$ contains the origin, corresponding to the standard conservation laws, and posssibly the centers of the facets of the Wigner–Seitz cell, corresponding to the staggered invariants.[28]

Both from a theoretical point of view and for the purpose of numerically evaluating (B.2) it is important to recognize that the two-particle propagator $[s(\mathbf{q})\,\omega]_{ij, kl} = \Gamma_{ik}(\mathbf{q}) \, \Gamma_{jl}(-\mathbf{q})$ can be written as the product of two single-particle propagators,

$$\Gamma_{ik}(\mathbf{q}) = e^{-i\mathbf{q} \cdot \mathbf{c}_i}\{\delta_{ik} + \Omega_{ik}^{(1, 1)}\} \qquad \text{(B.3)}$$

with biorthogonal right and left eigenvectors $\tilde{\psi}_\mu(\mathbf{q})$ and $\psi_\mu(\mathbf{q})$, and eigenvalues $\exp[z_\mu(\mathbf{q})]$. Each value of $\alpha = \mu\nu$ in (B.2) corresponds to a product mode with $\tilde{\chi}_{\alpha, ij}(\mathbf{q}) = \tilde{\psi}_{\mu, i}(\mathbf{q}) \, \tilde{\psi}_{\nu, j}(\mathbf{q})$, $\chi_{\alpha, ij}(\mathbf{q}) = \psi_{\mu, i}(\mathbf{q}) \, \psi_{\nu, j}(\mathbf{q})$, and $\lambda_\alpha(\mathbf{q}) = \exp[z_\mu(\mathbf{q}) + z_\nu(\mathbf{q})]$. The *slow* modes $\psi_{s, i}(\mathbf{q})$ of the Boltzmann propagator $\Gamma_{ik}(\mathbf{q})$ are related to the local conservation laws and have $z_s(0) = 0$. They

correspond to the zero left eigenvectors $\tilde{\psi}_s(0)$ of the linearized Boltzmann operator $\Omega^{(1,1)}$, which are linear combinations of the collisional invariants a_i. Products of slow modes at $\mathbf{q}=0$ give rise to $\lambda_\alpha = \exp[z_\mu(0) + z_\nu(0)] = 1$. Staggered modes have $z_\mu(\mathbf{q}_n) = z_\nu(\mathbf{q}_n) = i\pi$.

We can now formulate a necessary *solubility condition*, which requires that the inhomogeneous term in (3.8) be orthogonal to the null space, i.e.,

$$\langle \chi_\alpha(\mathbf{q}_n) \mid s(\mathbf{q}_n)B \rangle = 0 \tag{B.4}$$

for the modes (α, n) that have $\lambda_\alpha(\mathbf{q}_n) = 1$. It can be verified that this condition is indeed satisfied as a consequence of (2.25), i.e., the local conservation laws, since both for the standard conserved quantities $\mathcal{H}$ and for the staggered invariants we have $s(\mathbf{q}_n) = 1$.

After establishing that (3.8) is soluble, we observe that its solution is *not unique* in the set of points $\{\mathbf{q}_n\}$, since we may add an arbitrary linear combination of right null eigenvectors $\tilde{\chi}(\mathbf{q}_n)$ to a special solution of (B.1). Using $s(\mathbf{q}_n) = 1$, we have

$$\mathscr{g}_{ij}(\mathbf{q}_n) = \sum_{\alpha \notin \mathcal{N}_n} \tilde{\chi}_{\alpha,ij}(\mathbf{q}_n) \frac{1}{1 - \lambda_\alpha(\mathbf{q}_n)} \langle \chi_\alpha(\mathbf{q}) \mid B \rangle + \sum_{\alpha \in \mathcal{N}_n} \mathscr{E}_{n\alpha} \tilde{\chi}_{\alpha,ij}(\mathbf{q}_n) \tag{B.5}$$

It turns out that the coefficients $\mathscr{E}_{n\alpha}$ are completely determined by the covariance of the fluctuations of the (standard and spurious) global invariants. For instance, for the standard invariants $\mathcal{H} = \{N, \mathbf{P}, ...\}$ associated with $\mathbf{q}_n = 0$, the eigenmode $\chi_\alpha(0) = a_\lambda a_\mu$ is a product of collisional invariants. Using the biorthogonality relation $\langle \chi_\alpha \mid \tilde{\chi}_\beta \rangle = \delta_{\alpha\beta}$ we obtain from (B.5)

$$\mathscr{E}_{n\alpha} = \langle a_\lambda a_\mu \mid \hat{\mathscr{G}}_{ij}(0) \rangle = \sum_{\mathbf{r}} \sum_{ij} a_{\lambda,i} a_{\mu,j} \mathscr{g}_{ij}(\mathbf{r}) = \frac{1}{V} \langle \delta\mathcal{H}_\lambda \, \delta\mathcal{H}_\mu \rangle \tag{B.6}$$

The last equality follows from (2.15). In this paper we only consider the case where the initial ensemble is prepared *microcanonically*, i.e., with all global invariants fixed, so that all $\mathscr{E}_{n\alpha} = 0$.

Using the method of Section 3.1 we can again derive a linear equation of the form (3.17) for the excess on-node correlations $\mathscr{C}_{ij}$, defined by (3.15). The ring operator is now given by

$$R = \frac{1}{V} \sum_{\mathbf{q} \notin \{\mathbf{q}_n\}} \frac{1}{1 - s(\mathbf{q})\omega} s(\mathbf{q}) + \frac{1}{V} \sum_n \sum_{\alpha \notin \mathcal{N}_n} \tilde{\chi}_\alpha(\mathbf{q}_n) \frac{1}{1 - \lambda_\alpha(\mathbf{q}_n)} \chi_\alpha(\mathbf{q}_n) \tag{B.7}$$

Note that in the thermodynamic limit the contribution of the second term on the right-hand side is $\mathcal{O}(1/V)$, and therefore (B.7) reduces to (3.12).

For finite systems $\mathscr{G}_d \neq R(1-\omega)\,\mathscr{G}_d$, and as a consequence $\mathscr{G}_d$ contributes to the excess correlations $\mathscr{C}_{ij}$. These are given by (3.19), with M given by (3.18) as before, but with $K = R\Omega^{(2,\,0)} + J$ containing a finite-size correction term, $J \equiv R(1-\omega)\,\mathscr{G}_d - \mathscr{G}_d$, which is explicitly given by

$$J_{(ij)} = -\frac{1}{V} \sum_n \sum_{\alpha \in \mathcal{N}_n} \tilde{\chi}_{\alpha,\,ij}(\mathbf{q}_n)\langle \tilde{\chi}_\alpha(\mathbf{q}_n) \mid \mathscr{G}_d \rangle \qquad \text{(B.8)}$$

In deriving this equality we have used the fact that $s(\mathbf{q})\mathscr{G}_d = \mathscr{G}_d$, since $s_{ii}(\mathbf{q}) = 1$. The presence of this J term, together with the fact that expression (B.7) for the ring operator must be used instead of (3.12), accounts in a quantitative manner for finite-size effects of $\mathcal{O}(1/V)$.

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