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Nonequilibrium Steady States of Stochastic Lattice Gas Models of Fast Ionic Conductors

Sheldon Katz,[1,2] Joel L. Lebowitz,[1] and Herbert Spohn[1,3]

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We investigate theoretically and via computer simulation the stationary nonequilibrium states of a stochastic lattice gas under the influence of a uniform external field E. The effect of the field is to bias jumps in the field direction and thus produce a current carrying steady state. Simulations on a periodic 30×30 square lattice with attractive nearest-neighbor interactions suggest a nonequilibrium phase transition from a disordered phase to an ordered one, similar to the para-to-ferromagnetic transition in equilibrium $E = 0$. At low temperatures and large E the system segregates into two phases with an interface oriented parallel to the field. The critical temperature is larger than the equilibrium Onsager value at $E = 0$ and increases with the field. For repulsive interactions the usual equilibrium phase transition (ordering on sublattices) is suppressed. We report on conductivity, bulk diffusivity, structure function, etc. in the steady state over a wide range of temperature and electric field. We also present rigorous proofs of the Kubo formula for bulk diffusivity and electrical conductivity and show the positivity of the entropy production for a general class of stochastic lattice gases in a uniform electric field.

KEY WORDS: Steady states; stochastic lattice gas models; fast ionic conductors.

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[1] Department of Mathematics and Physics, Rutgers University, New Brunswick, New Jersey 08903.

[2] Present and permanent address: Department of Physics, Villanova University, Villanova, Pennsylvania. Work supported in part by a Lafayette College Junior Faculty Leave Grant.

[3] Present and permanent address: Theoretische Physik, Theresienstrasse 37, 8 Munchen 2, Germany. Work supported in part by a Heisenberg fellowship of the Deutsche Forschungsgemeinschaft.

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INTRODUCTION

The equilibrium properties of macroscopic systems, with a given microscopic Hamiltonian, can be obtained as suitable averages in well-defined Gibbs ensembles. While this only solves the problem in principle—the evaluation of integrals involving approximately 10^{23} variables is not simple—it does provide a starting point for both qualitative understanding and quantitative approximations to equilibrium behavior. In addition many interesting equilibrium phenomena, such as phase transitions, can be studied explicitly in simplified lattice model systems.

Our understanding of the statistical mechanics of nonequilibrium phenomena is much less satisfactory at the present time. We do not have any general prescription for choosing appropriate ensembles even for the simplest nonequilibrium systems, e.g., those in a stationary state in which steady particle or heat currents are maintained. Attempts to model such systems have so far yielded more or less explicit stationary ensembles only for noninteracting systems, e.g., ideal gases and harmonic crystals.[1]

In this note we describe theoretical investigations and computer simulations of the stationary nonequilibrium states of an interacting system often used to model a fast ionic conductor.[2] The nonequilibrium state is produced by a uniform external electric field E, which causes a steady current to flow in the system: the system may be thought of as a closed loop, represented by periodic boundary conditions. The system also interacts, stochastically, with a heat reservoir at a fixed temperature T. The reservoir absorbs the heat generated by the current and maintains the steady state.

In this idealized model system the particles or ions are located at lattice sites and interact with a nearest-neighbor pair potential J. Their stochastic dynamics consists of jumps to neighboring unoccupied sites (with periodic boundary conditions), i.e., Kawasaki-type dynamics. In the absence of an electric field, $E = 0$, the transition rates are such that the stationary state of the system is the Gibbs equilibrium state at the reservoir temperature T. The uniform field E, which is *not* the gradient of a potential, biases the jumps in the direction of E. (We may think of the field as produced by a uniformly increasing magnetic flux through a loop.)

Our rigorous results for this system are—despite the simplicity of the model—confined to rather general properties. We prove that, as expected, the total entropy production is positive and show that to *linear* order in E the steady state and the state of minimum entropy production coincide. We prove the validity of the Kubo formula and the Einstein relation for the electrical conductivity and bulk diffusivity at zero field. The Kubo formula for the bulk diffusivity also holds in the steady state with finite field E.

(This is, as far as we know, the first rigorous proof of the Kubo relation for a steady-state transport coefficient in an interacting system.)

The computer simulations on the other hand give more detailed if less reliable (certainly less rigorous) results. They were done on a half-filled one-dimensional lattice with 200 sites and on a two-dimensional square lattice of 900 sites—both with periodic boundary conditions. The results in two dimensions suggest the existence of a phase transition at low temperatures which is strongly influenced, perhaps even dominated, by the electric field. This is indicated by the apparent segregation of the system in a direction perpendicular to the field into a "fluid" and "gas" phase. For the equilibrium system, $E = 0$, a segregation into "randomly oriented" regions takes place (in a macroscopic system) below the Onsagar critical temperature $T_c \cong 0.44|J|/K_B$.[3] Defining the corresponding $T_c(E)$ as the apparent critical temperature in presence of the field we find that it increases with E for $J > 0$ (attractive) and decreases, possibly to zero, when E is very large, for $J < 0$ (repulsive). A rough phase diagram for $J > 0$, $E = \infty$, can thus be obtained. A preliminary account of our results has been given in Ref. 4.

In Section 1 we describe our model in more detail. In Sections 2 and 3 we provide some theoretical background for the numerical results presented in the following two sections. We report results on the steady-state current, the zero-frequency conductivity and bulk diffusivity, the structure function, the nearest-neighbor correlations, and the specific heat. The density of the system is always $1/2$ but we cover a wide range of temperatures and electric fields. The last three sections describe the exact results mentioned earlier and also include a discussion on the various ways used to determine numerically the bulk diffusivity. Some detailed calculations are left for the appendices.

1. DESCRIPTION OF THE MODEL AND DYNAMICS

We consider a stochastic lattice gas in the presence of a constant external electric field. Particles move on a simple (hyper) cubic lattice of dimension d. There is at most one particle per lattice site (hard-core exclusion). The system is confined to a box $\Lambda \subset \mathbb{Z}^d$ with periodic boundary conditions. The state space of the system is the set of all possible configurations. We denote the occupation variables by η_x,

$$\eta_x = \begin{cases} 1 & \text{if site } x \in \Lambda \text{ is occupied} \\ 0 & \text{is site } x \in \Lambda \text{ is empty} \end{cases}$$

and the full particle configuration by $\eta = \{\eta_x\}_{x \in \Lambda}$.

The dynamics is completely specified by prescribing the jump rates of the particles. To incorporate the hard-core exclusion it is convenient to think of them as exchange rates and to define

$$c(y,x,\eta) \equiv c(x,y,\eta) \geqslant 0 \tag{1.1}$$

as the rate at which the occupation at sites x and y are exchanged when the configuration is η. If $\eta_x = \eta_y$, then the exchange does not alter the configuration of the particles, considered identical here, and we may fix the value of $c(x,y,\eta)$ arbitrarily, e.g., equal to zero. (The situation would be different if we were to consider the motion of test particles, i.e., self-diffusion.[5,6]) If $\eta_x \neq \eta_y$, e.g., if $\eta_x = 1$ and $\eta_y = 0$, then $c(x,y,\eta)$ is the rate at which the particle at x jumps to the empty site at y. Letting η^{xy} denote the configuration η with occupation at sites x and y interchanged, i.e.,

$$(\eta^{xy})_z = \begin{cases} \eta_y & \text{for } z = x \\ \eta_x & \text{for } z = y \\ \eta_z & \text{for } z \neq x, y \end{cases}$$

the time evolution of a probability distribution $\rho_t(\eta)$ is given by the master equation

$$\begin{aligned} \frac{d}{dt}\rho_t(\eta) &= \frac{1}{2}\sum_{x,y\in\Lambda}\{c(x,y,\eta^{xy})\rho_t(\eta^{xy}) - c(x,y,\eta)\rho_t(\eta)\} \\ &= L\rho_t(\eta) \end{aligned} \tag{1.2}$$

To proceed we have to specify the jump rates. General considerations provide us with certain constraints, but as will be seen later some important properties of the model may depend on the detailed choice of the rates. We list and motivate our conditions on the jump rates $c_E(x,y,\eta)$. (We have added the subscript E to make the dependence on the electric field explicit.)

(i) For simplicity we allow only nearest-neighbor jumps, i.e., $c_E(x,y,\eta) = 0$ for $|x-y| > 1$. To avoid degeneracies it is also useful to assume that $c_E(x,y,\eta) > 0$ for $|x-y| = 1$ and $\eta_x \neq \eta_y$. This condition assures that the number of particles is the only conserved quantity. We will further assume, as is usually done, that $c_E(x,y,\eta)$ depends only on the $4d-2$ nearest neighbors of the bond (x,y).

(ii) Our model should describe correctly thermal equilibrium. Therefore for zero electric field we impose the condition of detailed balance

$$c_0(x,y,\eta) = c_0(x,y,\eta^{xy})e^{-\beta[H(\eta^{xy})-H(\eta)]} \tag{1.3}$$

Here β is the inverse temperature and $H(\eta)$ is the energy of the configura-

tion η. This we choose to be given by the usual nearest-neighbor Hamiltonian

$$H(\eta) = -\tfrac{1}{2}J \sum_{x,y\in\Lambda,\,|x-y|=1} \eta_x\eta_y \tag{1.4}$$

$J > 0$ corresponds to an attractive and $J < 0$ to a repulsive interaction.

To model the electric field one assumes that jumps in the direction of the field are favored and jumps opposite to the direction of the field are suppressed. Physically it seems reasonable still to assume a local detailed balance but now including the work done by the electric field in the jump. This yields

$$c_E(x, y, \eta) = c_E(x, y, \eta^{xy}) e^{-\beta[H(\eta^{xy}) - H(\eta)]} e^{E\cdot(x-y)(\eta_x - \eta_y)} \tag{1.5}$$

Physically E stands for the combination $q\beta E$, where q is the charge and E the applied external electric field.

Remark. Notice that because of the periodic boundary conditions the electric field energy cannot be included in the Hamiltonian. In particular

$$Z^{-1}\exp\left[-\beta H(\eta) + \sum_x E\cdot x\eta_x\right] \tag{1.6}$$

is *not* invariant under the dynamics with rates $c_E(x, y, \eta)$, i.e., it is not a stationary solution of (1.2). For boundary conditions corresponding to a closed box (1.6) would be invariant. This state would describe then a gas in a uniform field in thermal equilibrium with no transport, which is clearly of no interest to us here.

(iii) Finally, it seems natural, although not necessary, to assume certain symmetry and homogeneity properties of the jump rates. We require that the rates are invariant under translations, i.e., $c_E(x + a, y + a, \tau_a\eta) = c_E(x, y, \eta)$ where $\tau_a\eta$ denotes the configuration η shifted by a, modulo the periodicity of Λ. We also require that the rates are left invariant under a reflection of both the configuration and the E-field. Lastly if we think of particles as positive charges and empty sites as negative charges, then it is physically natural to assume symmetry under charge conjugation. This means, in lattice gas language, that if we change η_x to $1 - \eta_x$ for all x and E to $-E$ then the rates remain unchanged.

There is a simple way to implement all these symmetries through setting

$$c(x, y, \eta) = \phi(\beta H(\eta^{xy}) - \beta H(\eta) - E\cdot(x-y)(\eta_x - \eta_y)) \tag{1.7}$$

with some function ϕ which, by detailed balance (1.5) has to satisfy $\phi(h) = \phi(-h)e^{-h}$. In principle ϕ could still depend on $|E|$ and β; in fact

the rate of hopping is strongly temperature dependent. However, it seems reasonable to assume that this dependence enters ϕ only multiplicatively and that otherwise ϕ is independent of β and E. Since a change in time scale does not affect the steady state, our prime interest here, we may as well assume that ϕ is independent of β and E. In particular we may adopt then one of the well-known choices for ϕ at $E = 0$: for the Metropolis dynamics[7] $\phi(h) = 1$ for $h \leqslant 0$ and $\phi(h) = e^{-h}$ for $h \geqslant 0$ and for the Kawasaki dynamics[8] $\phi(h) = 1/(1 + e^h)$.

For simplicity we shall absorb the coupling constant into the inverse temperature and set βJ equal to 4β. Then $\beta > 0$ corresponds to an attractive interaction, $\beta < 0$ to a repulsive interaction, and $\beta = 0$ to a noninteracting system.

2. STATIONARY STATES

We are interested in the steady nonequilibrium situation and investigate here general properties of stationary states. A stationary probability distribution $\rho_{E,N}$ is defined by

$$L_E \rho_{E,N}(\eta) = 0 \tag{2.1}$$

Because the number N of particles is conserved we have a stationary distribution for each $N = 0, \ldots, |\Lambda|$ and since the jump rates are nondegenerate these stationary distributions are unique. Expectations with respect to $\rho_{E,N}$ are denoted by $\langle \cdot \rangle_{E,\rho}$, where $\rho = N/|\Lambda|$. $\rho_{0,N}$ is the canonical equilibrium state.

The theory of Markov chains ensures an exponentially fast approach to stationarity for finite Λ. However, because of the conservation law, the approach to stationarity is diffusive and therefore slow. The time needed to approach stationarity is estimated to be of the same order as that required for a random walk in Λ to reach its equilibrium. Thus the first nonzero eigenvalue of L_E is expected to be of the order $|\Lambda|^{-2}$. At low temperatures the approach to stationarity may be even slower.

In contrast to the thermal equilibrium situation, $E = 0$, $\rho_{E,N}$ is defined only through (2.1) and not known explicitly, except for the few cases mentioned below. We outline briefly our information about $\rho_{E,N}$:

$\rho_{E,N}$ has to inherit the symmetries of the rates. Therefore $\rho_{E,N}$ is invariant under translations, i.e.,

$$\rho_{E,N}(\eta) = \rho_{E,N}(\tau_a \eta) \tag{2.2}$$

If for some reflection we denote by $R\eta$ the reflected configuration η and by RE the correspondingly reflected field, then

$$\rho_{E,N}(\eta) = \rho_{RE,N}(R\eta) \tag{2.3}$$

If we denote by $C\eta$ the configuration η with particles and empty sites interchanged, then

$$\rho_{E,N}(\eta) = \rho_{-E,|\Lambda|-N}(C\eta) \tag{2.4}$$

The symmetry of the probability distribution implies of course the corresponding symmetry of expectations. For example, (2.3) implies that the average current is odd in E, and together with (2.4) we conclude that the average current is even in the density around $\rho = 1/2$.

We note that in general $\rho_{E,N}$ is *not* even in E. Higher-order correlations which cannot be related to each other by reflection distinguish between the states $\rho_{E,N}$ and $\rho_{-E,N}$.

In a few cases (2.1) can be solved explicitly. For the particular choice

$$c_E(x, y, \eta) = e^{E\cdot(x-y)(\eta_x-\eta_y)/2} \tag{2.5}$$

which corresponds to $\beta = 0$ and $\phi(h) = e^{-h/2}$ in (1.7), the invariant distribution $\rho_{E,N}$ is obtained for arbitrary dimension by randomly placing N particles in Λ. The invariant distribution does not depend on E and corresponds to the equilibrium state at infinite temperature. Notice, however, that merely setting $\beta = 0$ in (1.5) does not ensure that the stationary state is of that simple form.

In one dimension there is a whole class of nontrivial jump rates, cf. Appendix 4, for which the canonical equilibrium state $Z^{-1}e^{-\beta H(\eta)}$ with N particles is invariant under L_E independently of the magnitude of E. Therefore all quantities of interest such as steady-state current, structure function, can be computed explicitly. In two or more dimensions, with the exception of (2.5), it is impossible to choose the jump rates in such a way that the thermal equilibrium state remains invariant as the E field is turned on.

All explicit solutions have the feature that the steady state is independent of E. Except for a few particles on a ring[9] we have no example with a nontrivial E dependence.

3. STEADY STATE CURRENT, ZERO FIELD CONDUCTIVITY

A quantity of basic physical interest is the average current in the steady state. We give first some theoretical background and describe in the following section our numerical results.

Let us define the current function $j_E(x, y)$ for the bond (x, y) by

$$j_E(x, y)(\eta) = c_E(x, y, \eta)(\eta_x - \eta_y) \tag{3.1}$$

This is the expected jump rate from x to y in a configuration η. The average

current in the mth coordinate direction is then given by

$$j_m(\beta, E) = \langle j_E(x, x + e_m)\rangle_{E,\rho} \tag{3.2}$$

Here e_m is the unit vector pointing along the positive m axis. By translation invariance the average does not depend on x and by reflection symmetry it is odd in E.

The zero field conductivity is given by

$$\sigma_{mn} = \frac{\partial}{\partial E_n} \langle j_E(x, x + e_m)\rangle_{E,\rho}|_{E=0} \tag{3.3}$$

Let us denote the differentiation in (3.3) by a prime. Then

$$\begin{aligned}\sigma_{mn} = &\sum_{\eta} \rho_{0,N}(\eta) c_0'(x, x + e_m, \eta)(\eta_x - \eta_{x+e_m}) \\ &+ \sum_{\eta} \rho_{0,N}'(\eta) c_0(x, x + e_m, \eta)(\eta_x - \eta_{x+e_m})\end{aligned} \tag{3.4}$$

Here $\rho_{0,N}$ is the canonical equilibrium state. Note that we have two contributions: the first one results because of the E dependence of the current function and the second one because of the E dependence of the steady state. Using the detailed balance condition (1.5), cf. Appendix 1 for the computational details, these two terms correspond, respectively, to the static and dynamic part of the conductivity

$$\begin{aligned}\sigma_{mn} = &\tfrac{1}{2}\delta_{mn}\langle c_0(0, e_n, \eta)(\eta_0 - \eta_{e_n})^2\rangle_{0,\rho} \\ &- \int_0^\infty dt \sum_x \langle j_0(x, x + e_n) e^{L_0^* t} j_0(0, e_m)\rangle_{0,\rho}\end{aligned} \tag{3.5}$$

The expectations and rates are the zero field equilibrium expectations and rates. L_0^* is the adjoint of L_0 defined in (1.2). By reflection symmetry the second contribution in (3.5) and therefore the conductivity tensor are diagonal.

Using the same reasoning it can be shown that the frequency-dependent conductivity $\sigma_{mn}(\omega)$ is also of the form (3.5) with the addition of the factor $e^{i\omega t}$ under the time integral. Thus only the change in the steady state, which results from applying the electric field, leads to a frequency dependence of the conductivity. In particular, in all exactly soluble examples mentioned above, the steady state does not depend on E and therefore $\sigma_{mn}(\omega)$ is frequency independent.

We will show in Section 6 that (3.5) is really the Kubo formula for the conductivity. It is not immediately obvious that this is in fact what (3.5) is (note in particular that the second term is negative), but this is indeed the case. Just as for Hamiltonian systems, the Kubo formula here expresses the transport coefficient as the space-time integral over the corresponding

current–current correlation function. It is the definition of the appropriate current function which requires some care in the stochastic case.

Since by detailed balance (1.3) $e^{L_0^* t}$ is symmetric with respect to the equilibrium weight $Z^{-1}e^{-\beta H(\eta)}$, and L_0^* is negative, we have the spectral representation

$$\int_0^\infty dt \sum_x \langle j_0(x, x+e_n) e^{L_0^* t} j_0(0, e_m)\rangle_{0,\rho}$$

$$= \delta_{mn} \int_0^\infty dt \int_0^\infty \mu(d\lambda)\, e^{-\lambda t} = \delta_{mn} \int_0^\infty \lambda^{-1} \mu(d\lambda) \tag{3.6}$$

with some positive measure $\mu(d\lambda)$. This yields immediately the upper bound

$$\sigma_{mn} \leqslant \tfrac{1}{2}\delta_{mn} \langle c_0(0, e_n, \eta)(\eta_0 - \eta_{0_n})^2\rangle_{0,\rho} \tag{3.7}$$

As noticed by Halperin,[10] (3.7) can be improved by Jensen's inequality to give

$$\sigma_{mn} \leqslant \tfrac{1}{2}\delta_{mn} \langle c_0(0, e_n, \eta)(\eta_0 - \eta_{e_n})^2\rangle_{0,\rho} - \left[\int_0^\infty \mu(d\lambda)\right]^2 \Big/ \int_0^\infty \mu(d\lambda)\lambda$$

$$= \tfrac{1}{2}\delta_{mn} \langle c_0(0, e_n, \eta)(\eta_0 - \eta_{e_n})^2\rangle_{0,\rho}$$

$$- \left[\sum_x \langle j_0(x, x+e_n) j_0(0, e_n)\rangle_{0,\rho}\right]^2 \Big/ \sum_x \langle j_0(x, x+e_n) L_0^* j_0(0, e_n)\rangle_{0,\rho} \tag{3.8}$$

The Mori expansion (continued fraction expansion)[11] goes beyond (3.8). One assumes that some moments of the spectral measure $\mu(d\lambda)$, i.e.,

$$\int_0^\infty \mu(d\lambda)\lambda^k = \sum_x \langle j_0(x, x+e_n)(L_0^*)^k j_0(0, e_n)\rangle_{0,\rho} \tag{3.9}$$

have been computed for $k = 0, 1, \ldots, 2M-1$. Then $\mu(d\lambda)$ is taken to be the sum of M delta functions fitted to yield the moments (3.9) and one evaluates on this basis $\int_0^\infty \mu(d\lambda)\, 1/\lambda$. The Mori expansion is based on the assumption that the spectral distribution is dominated by a few frequencies. As will be discussed in Section 4, we checked this assumption for the one-dimensional model and found it not to be valid in this case.

4. NUMERICAL RESULTS I: STEADY STATE CURRENT, CONDUCTIVITY

For our numerical study we used in one dimension a system of 200 lattice sites with 100 particles and in two dimensions a 30×30 square lattice with 450 particles. Since we always kept the density equal to $1/2$, we do not indicate the density dependence. We use standard Monte Carlo

procedure. A bond (x, y) is randomly selected. If $\eta_x = \eta_y$ a new bond is randomly selected. If $\eta_x \neq \eta_y$ then in two dimensions if in the attempted exchange the energy difference including the work done by the electric field, ΔU, is negative the exchange is performed, and if ΔU is positive, then the exchange is performed with probability $e^{-\beta \Delta U}$. This choice corresponds to setting $\phi(h) = 1$ for $h \leqslant 0$ and $\phi(h) = e^{-h}$ for $h \geqslant 0$ in (1.7). In one dimension we modified somewhat the Metropolis dynamics in order to still have a nontrivial E dependence of the steady state even at $\beta = 0$: we used the rates

$$\begin{aligned} &c_E(x, x+1; 0,1,0,0) = 1, \qquad c_E(x, x+1; 0,0,1,0) = e^{-E} \\ &c_E(x, x+1; 1,1,0,1) = 1, \qquad c_E(x, x+1; 1,0,1,1) = e^{-E} \\ &c_E(x, x+1; 1,1,0,0) = \tfrac{1}{2}\min(1, e^{-4\beta}), \\ &c_E(x, x+1; 0,0,1,1) = \tfrac{1}{2}e^{-E}\min(1, e^{-4\beta}) \\ &c_E(x, x+1; 0,1,0,1) = \tfrac{1}{2}\min(1, e^{4\beta}), \\ &c_E(x, x+1; 1,0,1,0) = \tfrac{1}{2}e^{-E}\min(1, e^{4\beta}) \end{aligned} \tag{4.1}$$

They are still of the form (1.7) but with ϕ depending on E.

The natural time scale is calibrated in Monte Carlo step per site (MCS) units. One MCS is 900 (200) attempted exchanges in two (one) dimensions. The time scale of the master equation (1.2) is in MCS units. The particles start in a random configuration and are then quenched to the desired temperature at $E = 0$. The system evolves for 15000 MCS to ensure a typical equilibrium configuration. This configuration is saved for the following runs. Then the E field is gradually turned on. We allow for another 15000 MCS to ensure a typical steady-state configuration. Quantities such as the current and correlation functions were calculated by averaging over 25000 MCS in a given run. The data quoted are an average over at least six runs for $d = 1$ (two to five runs for $d = 2$) at a given temperature and electric field.

To obtain the steady-state current in the mth direction one counts the total number, $N_{+,m}(t)$, of jumps in the direction of e_m and the total number, $N_{-,m}(t)$, of jumps in the direction of $-e_m$ during a time interval of length t. The current is then

$$j_m(\beta, E) = \frac{1}{|\Lambda| t}\left[N_{+,m}(t) - N_{-,m}(t)\right], \qquad t \gg 1 \tag{4.2}$$

Also of interest is the number of exchanges per bond per unit time numerically defined by

$$w_m(\beta, E) = \frac{1}{|\Lambda| t}\left[N_{+,m}(t) + N_{-,m}(t)\right], \qquad t \gg 1 \tag{4.3}$$

This quantity is given by

$$w_m(\beta, E) = \langle c_E(x, x + e_m, \eta)(\eta_x - \eta_{x+e_m})^2 \rangle_{E,\rho} \tag{4.4}$$

$w_m(\beta, E)$ measures how often an exchange is actually performed. In thermal equilibrium $j_m(\beta, 0) = 0$. If the E field points in the mth direction, then for large fields $N_{+,m}(t) \gg N_{-,m}(t)$ and $j_m(\beta, E) \cong w_m(\beta, E)$. This defines then the region of saturation. Any further increase in the E field no longer changes the steady state. In Fig. 1 we show an example with $d = 1$ and $\beta = 0.6$. Since E enters in the exponential, $E = 5$ means that jumps opposite to the field are suppressed by a factor 7×10^{-3}. Numerically this corresponds to $E = \infty$.

For given β the current increases first linearly in E and then reaches saturation as $E \to \infty$. For low temperatures and attractive interaction the current is strongly suppressed because of clustering. For repulsive interaction the current is also suppressed at low temperatures, but the phenomenon sets in more slowly. There is no symmetry under $\beta \to -\beta$. The maximum zero-field conductivity and maximum saturation current occur at small negative β. We fit our data to the empirical guess $a(1 - e^{-bE})$,

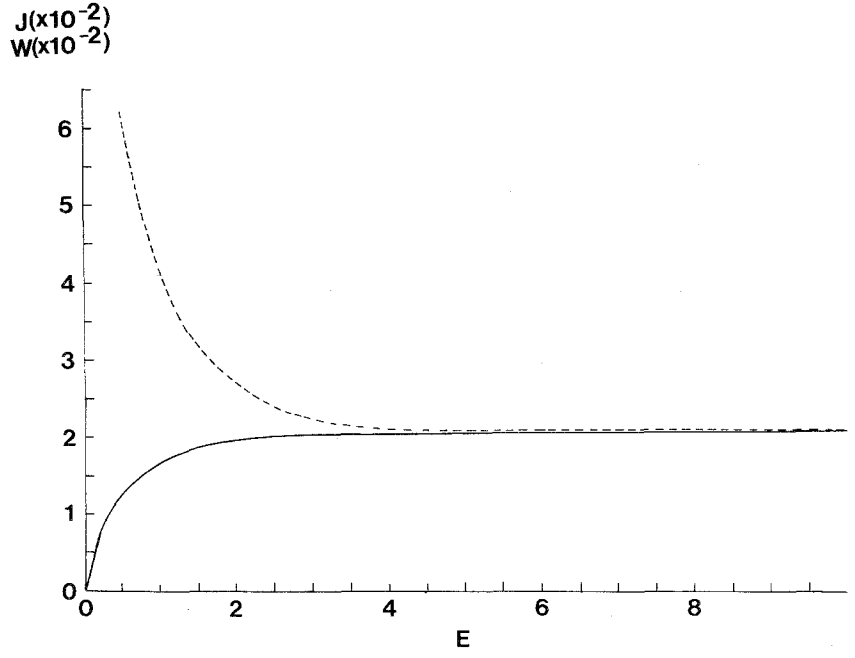
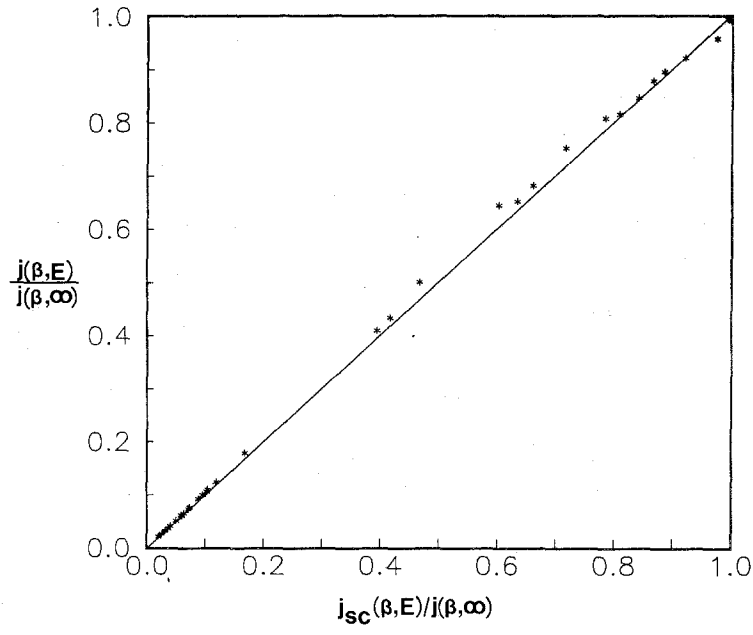
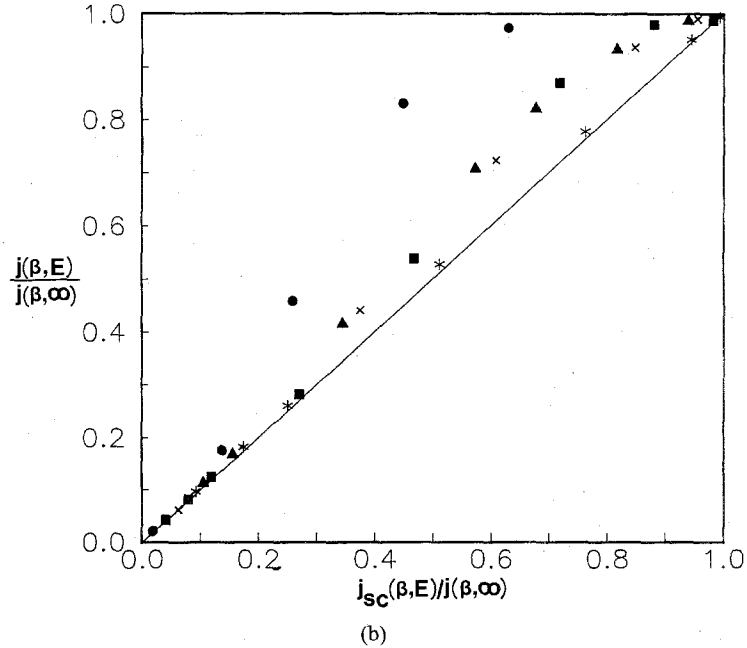


Fig. 1. Current (solid line) and average exchange rate (dotted line) as a function of electric field in one dimension at $\beta = 0.6$.



(a)



(b)

Fig. 2. (a) Scaling of the current in one dimension. Included are data for $\beta = -0.4, -0.2, 0.0, 0.2, 0.4, 0.6$ and several values of electric field from zero to saturation. (b) Scaling of the current in two dimensions. The asterisks correspond to $\beta = 0.0$, the full triangles to $\beta = 0.2$, the full squares to $\beta = 0.4$, the crosses to $\beta = -0.2$ and the full circles to $\beta = -0.4$.

$E \geqslant 0$, such that the zero-field conductivity, $(\partial/\partial E_m) j_m(\beta, 0) = \sigma(\beta, 0)$, and the saturation current are exact. This yields the scaling function

$$j_{sc}(\beta, E) = j(\beta, \infty)(1 - e^{-E\sigma(\beta,0)/j(\beta,\infty)}) \tag{4.5}$$

Figures 2a and 2b show that the data fit reasonably well in one dimension and in two dimensions at high temperatures. In all cases $j(\beta, E) \geqslant j_{sc}(\beta, E)$.

Physically one would expect that a strong E field should override any kind of thermal dependence of the steady state. The detailed balance condition by itself does not imply such a behavior for the simple reason that it gives no connection between different (β, E) values. But if we choose the rates to be of the form (1.7) with ϕ independent of β and E and with $\phi(h) \to \text{const}$ as $h \to -\infty$, then in one dimension as $|E| \to \infty$ the rates are up to a multiplicative factor of the form (2.5) and therefore the steady state is the infinite temperature equilibrium state *independently* of the choice of β. This is why we choose rates (4.1) which still contain a temperature dependence even at $E = \infty$. In two or more dimensions if the field is directed along one of the coordinate axes, but not otherwise, even rates of the form (1.7) still retain a nontrivial temperature dependence at $E = \infty$ because of the jumps orthogonal to the field which are still governed by the thermal rates. These facts show the strong dependence of the stationary states on the dynamics, which in a way reflects the rigidity of our model: particles can only be on lattice sites, the lattice is never distorted, jumps can take place only to nearest neighbors.

From the steady-state current we obtain directly the zero-field conductivity $\sigma(\beta, 0) \equiv \sigma(0)$. We list our numerical values in Table I.

According to (3.5) the zero-field conductivity has two contributions: a first one which is just one half of the average exchange rate, and a second one of dynamical origin. We follow the literature and denote the static contribution by $\sigma(\infty)$. As a measure for the dynamical contribution to the conductivity one defines the correlation factor

$$f_c = \frac{\sigma(0)}{\sigma(\infty)} \tag{4.6}$$

$f_c \leqslant 1$ by (3.7). By (4.2) and (4.3) f_c is numerically obtained as

$$f_c = \lim_{E \to 0} \frac{2}{E} \frac{N_+(t) - N_-(t)}{N_+(t) + N_-(t)} \tag{4.7}$$

with t large in order to have small statistical fluctuations. In Table I, we list the correlation factor at different temperatures in one and two dimensions, respectively. It is seen that the dynamical contribution to the zero-field conductivity is not negligible in general. In one dimension $f_c < 1$ even at

Table I[a]

(a)

β	$\sigma(\infty)$ theory	$\sigma(\infty)$	$\sigma(0)$	$\sigma(0)$ upper bound	f_c	$\int dt\langle j(t)j(0)\rangle$	one-pole	error (%)	D
−0.4	0.1808	0.181	0.148	0.161	0.82	0.033	0.0196	41	1.318
−0.2	0.1920	0.192	0.165	0.175	0.84	0.027	0.0168	38	1.146
0.0	0.1875	0.188	0.174	0.179	0.93	0.014	0.0083	41	0.696
0.2	0.1288	0.129	0.111	0.118	0.86	0.017	0.0108	36	0.298
0.4	0.0812	0.081	0.067	0.073	0.83	0.014	0.0083	41	0.146
0.6	0.0475	0.047	0.038	0.043	0.81	0.009	0.0047	48	0.057

(b)

β	$\sigma(\infty)$	$\sigma(0)$	f_c	D
−0.4	0.037	0.029	0.78	0.504
−0.2	0.091	0.088	0.96	0.694
0.0	0.125	0.125	1	0.500
0.2	0.065	0.059	0.91	0.091
0.4	0.024	0.019	0.79	0.002

[a] Part (a) refers to one dimension, with columns (1) $\sigma(\infty)$ the infinite frequency part of the conductivity, analytically computed from the first term of (3.5). (2) The same quantity numerically determined through the average exchange rate. (3) The static conductivity $\sigma(0)$. (4) The theoretical upper bound (3.8). (5) $f_c = \sigma(0)/\sigma(\infty)$. (6) The time integral in (3.5). (7) The same quantity in the one-pole approximation. (8) The relative error of the one-pole approximation. (9) The bulk diffusion coefficient determined from the Einstein relation $D = \sigma/\chi$. Part (b) refers to the same quantities in two dimensions.

$\beta = 0$. This originates in our particular choice (4.1). If in (4.1) $\frac{1}{2}\min(1, e^{\pm 4\beta})$ is replaced by $\min(1, e^{\pm 4\beta})$, then $f_c = 1$ at $\beta = 0$.

For one-dimensional models with arbitrary rates Singer[12,13] computed analytically, with the aid of a computer algorithm, the first five moments of the spectral measure $\mu(d\lambda)$, cf. (3.9), i.e.,

$$\sum_x \langle j_0(x, x+1, \eta)(L_0^*)^k j_0(0, 1, \eta)\rangle_{0,\rho} \tag{4.8}$$

for $k = 0, 1, \ldots, 5$. This yields then a theoretical value for the upper bound (3.8) which is listed in Table I, column 7. Singer evaluated the one-, two-, and three-pole approximation according to the Mori scheme. He found that the one-pole approximation differs only little from the three-pole one. From Table I, column 8, we see that the one-pole approximation is about 40% off over the whole temperature range, which indicates that the spectral distribution $\mu(d\lambda)$ is not sharply peaked.

For one-dimensional models Zeyher[14] and Singer[12] argue that the current–current correlation function has a long time tail, i.e.,

$$\int_0^\infty \mu(d\lambda)\, e^{-\lambda t} \sim t^{-3} \tag{4.9}$$

for long times. In our computer simulation the current–current correlation function decayed within 2MCS to the noise level over the whole temperature range. This would seem to indicate that any long time tail—if present—must have a very small coefficient.

As will be discussed in Section 6, the bulk diffusion coefficient D and the zero-field conductivity are related by the Einstein relation

$$D = \sigma/\chi \tag{4.10}$$

where χ is the compressibility,

$$\chi(\rho) = \sum_x \left(\langle \eta_x \eta_0 \rangle_{0,\rho} - \rho^2 \right) \tag{4.11}$$

We used series expansions for $\chi(\rho)$[15,16] and list in Table I, columns 9 and 13 our numerical values for the bulk diffusivity at density 1/2.

5. NUMERICAL RESULTS II: STRUCTURE FUNCTION, NEAREST-NEIGHBOR CORRELATIONS, LONG-RANGE ORDER, AND PHASE TRANSITION

A quantity of physical interest is the density–density correlation function in the steady state. Its Fourier transform defines the zero-frequency structure function $S(k)$ which is proportional to the differential scattering cross section. The small-k behavior of $S(k)$ yields information about the decay of correlations in the steady state and on the possible occurrence of long-range order.

We investigated numerically the structure function in one and two dimensions. The structure function is defined by

$$S_{\beta,E}(k) = \frac{1}{|\Lambda|} \sum_\eta \rho_{E,N}(\eta) \left| \sum_{x \in \Lambda} e^{ikx} \eta_x \right|^2 \tag{5.1}$$

k runs over the first Brillouin zone. In one dimension $N = |\Lambda|/2$ and $k = (2\pi/200)n$ with $n = 0, 1, \ldots, 199$. In two dimensions $k_1 = (2\pi/30)n_1$, $k_2 = (2\pi/30)n_2$ with $n_1, n_2 = 0, \ldots, 29$. For a repulsive interaction it is more instructive to study the staggered structure function $\tilde{S}_{\beta,E}(k)$ which is obtained by shifting in $S_{\beta,E}(k)$ each component of k by π. Numerically the steady-state average (5.1) is obtained by time averaging.

Since the results depend on the dimension, we discuss one and two dimensions separately.

$d = 1$, *Attractive Interaction.* For our choice of rates the E field acts like an increased ferromagnetic coupling. This is reflected in the structure function $S(k)$, cf. Fig. 3. The central peak increases with increasing field and the correlation length increases roughly by a factor of 4 from $E = 0$ to

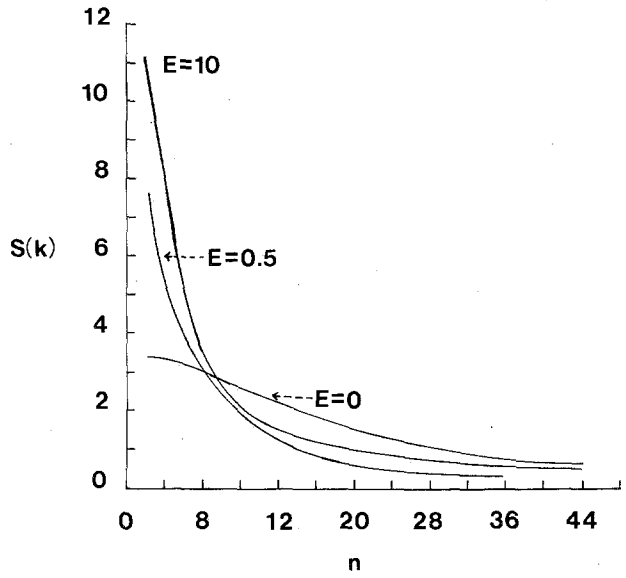


Fig. 3. The structure function $S(k)$ where $k = (2\pi/200)n$ for several values of electric field in one dimension at $\beta = 0.6$.

$E = \infty$. The "effective temperature," determined by fitting either to the equilibrium nearest-neighbor correlation or to the equilibrium correlation length, decreases roughly by a factor of $\frac{1}{2}$. The observed phenomenon cannot be of general nature. If we were to choose rates satisfying (A.26), then the correlation length would be independent of E, and if we were to choose the Metropolis dynamics, then for large fields the rates would become independent of β and the correlations would tend to zero as $E \to \infty$.

$d = 1$, *Repulsive Interaction.* Over the temperature range considered the structure function is practically independent of E.

$d = 2$, *Attractive Interaction.* In two dimensions we oriented the field always along the negative y axis. According to Onsager the equilibrium (infinite) system has a phase transition at $\beta_c \cong 0.44$. For $\beta > \beta_c$ there is a liquid (high density) and a gas (low density) phase. As $\beta \to \beta_c$, at $\rho = 1/2$, the correlation length diverges and the structure function becomes singular at $\mathbf{k} = 0$. This behavior is also reflected in the 30×30 system. Since the number of particles does not fluctuate, $S_{\beta,E}(\mathbf{k} = 0) = 0$ for all E and β. The maximum of $S_{\beta,0}(\mathbf{k})$ occurs at $\mathbf{k} = (2\pi/30, 0)$ and by symmetry also at $\mathbf{k} = (0, 2\pi/30)$. As we approach the region around 0.44 this maximum increases rapidly by an order of magnitude.

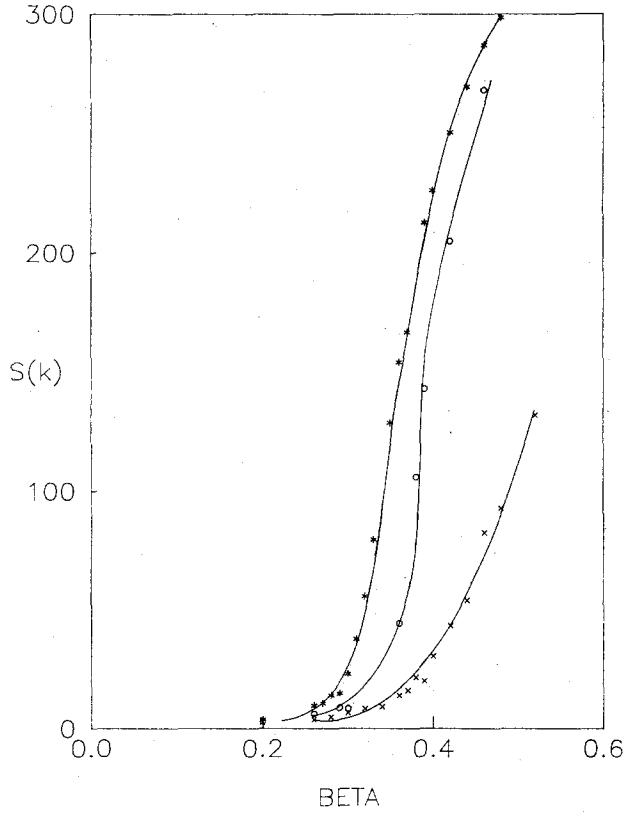
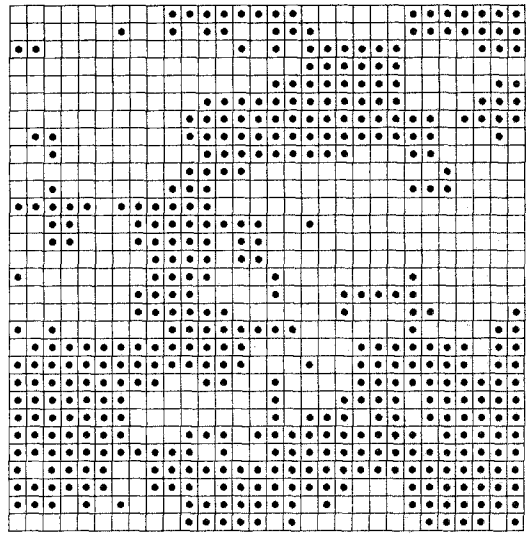
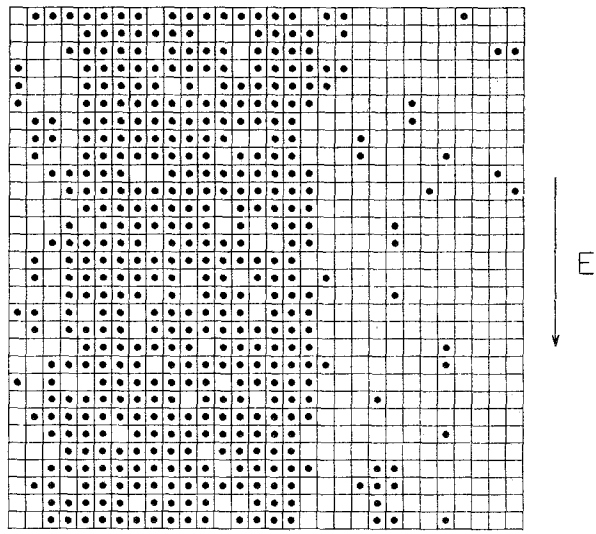


Fig. 4. The structure function $S(k)$ at the value $k_x = 2\pi/30$, $k_y = 0$, as a function of β, for various electric fields. The crosses correspond to $E = 0$, the open circles to $E = 0.75$, and the asterisks to $E = \infty$.

For finite E (always oriented along the negative y axis) we observe first that the region of rapid increase in the maximum of $S_{\beta,E}(\mathbf{k})$ occurs at smaller β, cf. Fig. 4. At $E = \infty$ the rapid increase occurs around $\beta = 0.32$. Secondly the structure function becomes highly anisotropic. While $S_{\beta,\infty}(0, 2\pi/30) \cong 1$ over the whole temperature range considered, $S_{\beta,\infty}(2\pi/30, 0)$ increases from 1 to about 225. This suggests defining a critical temperature $\beta_c(E)$ depending on E, as the value for which $S_{\beta_c(E),E}(2\pi/30, 0) \cong 50$ (the choice 50 being somewhat arbitrary of course). Studying typical configurations the following picture emerges: for $\beta < \beta_c(E)$ we have a disordered configuration with some tendency to form clusters whereas for $\beta > \beta_c(E)$ and $E \neq 0$ typical configurations form striplike clusters oriented parallel to the field. In Fig. 5 we display some

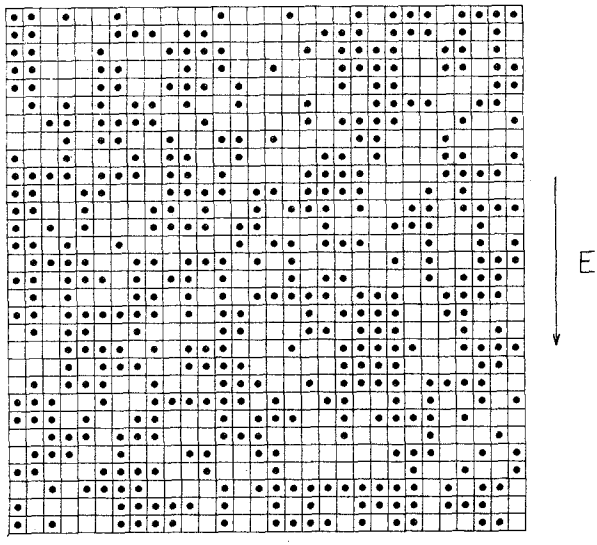


(a)



(b)

Fig. 5. (a) Equilibrium configuration at $\beta = 0.40$, $E = 0$. (b) A typical steady-state configuration, showing the formation of fully occupied columns at $\beta = 0.40$, and saturation field.



(c)

Fig. 5. (continued) (c) A typical steady-state configuration at $\beta = 0.20$ and saturation field.

Table II. Some Values of the Structure Function $S(K)$ for: (a) $E = 0$, $K = 0.4$, (b) $E = \infty$, $K = 0.4$, and (c) $E = \infty$, $K = 0.2$

	n_1	0	1	2	3	4	5	
n_2	0	0.0	27.4	8.8	4.3	2.9	1.9	
	1	33.8	16.2	8.3	4.3	3.1	2.1	
	2	9.7	6.8	5.0	2.9	2.0	1.6	(a)
	3	4.5	4.4	2.9	2.2	1.6	1.4	
	4	2.9	2.7	2.3	1.5	1.3	1.2	
	5	1.9	1.5	1.4	1.4	1.3	0.9	
	n_1	0	1	2	3	4	5	
n_2	0	0.0	226.2	1.5	18.6	1.6	4.4	
	1	0.7	0.7	1.0	0.9	1.2	1.1	
	2	0.5	0.5	0.7	0.7	0.7	0.9	
	3	0.4	0.4	0.5	0.5	0.6	0.7	(b)
	4	0.4	0.4	0.4	0.5	0.5	0.4	
	5	0.4	0.4	0.3	0.5	0.5	0.5	
	n_1	0	1	2	3	4	5	
n_2	0	0.0	3.8	3.4	3.4	2.4	2.2	
	1	0.9	1.2	2.2	2.9	2.4	2.3	
	3	1.0	1.2	1.7	2.0	1.8	2.2	(c)
	4	1.0	1.0	1.1	1.4	1.8	1.5	
	5	0.7	0.9	0.9	1.1	1.2	1.4	

typical configurations contrasting the cases $\beta < \beta_c(\infty)$ with $\beta > \beta_c(\infty)$ and $E = 0$ with $E = \infty$ for $\beta > \beta_c(\infty)$. As a more quantitative measure we also show the structure function for small k in Table II.

To give an explanation which is at least qualitative for the occurrence of strip configurations it is useful to consider very low temperatures. If $E = 0$, then jumps which decrease the number of nearest neighbors are essentially forbidden. The ground state is a droplet which has the smallest surface, and at low temperatures the system tries to minimize surface energy. On the other hand if the work done by the field is larger than the binding energy, say $E = \infty$, then jumps in the direction of the field still

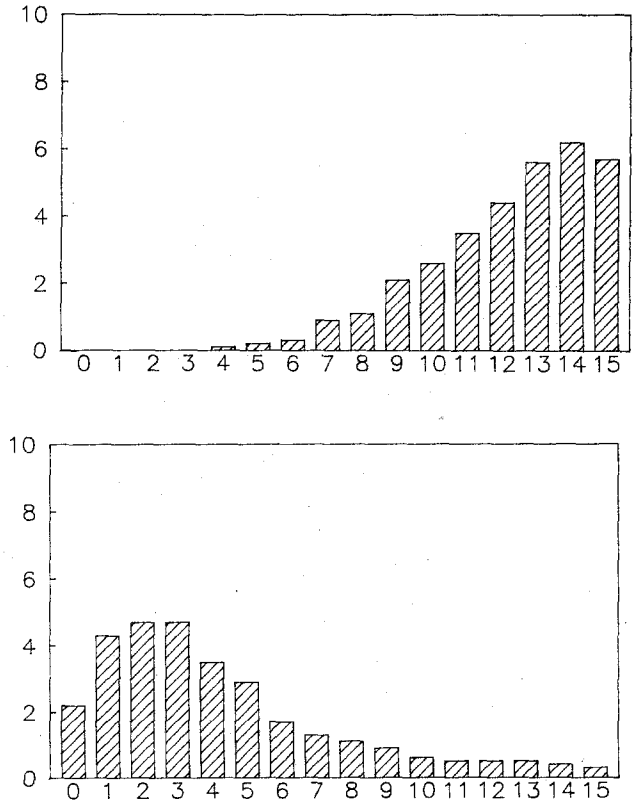


Fig. 6. Histograms showing the average number of columns (vertical axis) with a given number of occupied sites. Because of symmetry, columns with m occupied sites and $(30 - m)$ unoccupied sites are considered equivalent to columns with m unoccupied sites and $(30 - m)$ occupied sites. Because such symmetry does not exist for $m = 15$, contributions for $m = 15$ were multiplied by two in compiling the histogram. (a) Histogram from $\beta = 0.20$ and saturation field. (b) Histogram for $\beta = 0.40$ and saturation field.

have rate one independently of the number of nearest neighbors, provided the jump is not blocked. Therefore the most slowly changing configurations are strips parallel to the field. In analogy to thermal equilibrium we would expect that the transition from disordered to strip configurations becomes sharp for the infinite system.

We may learn something about the phase diagram in the β, ρ plane for large E ($E \cong \infty$) by looking at the densities of the "liquid" and "gas" phases as they appear in typical configurations at $\beta > \beta_c(\infty)$. Counting the number in each column we can make a histogram whose peaks, for very large systems, should occur at ρ_v and ρ_l, respectively. By the symmetries (2.3)–(2.4), $\rho_l - \rho_v = m^*$, where m^* is the spontaneous magnetization in spin language. Such histograms are shown in Fig. 6. To take account of the finite (small) size of our system we estimate m^* by computing the difference

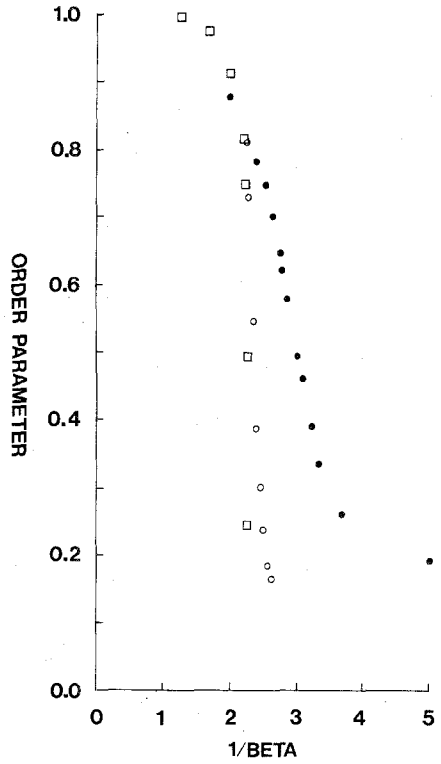


Fig. 7. Order parameter vs. $1/\beta$. The solid circles represent our Monte Carlo data in maximum electric field. The open squares represent the exact zero electric field magnetization and the open circles represent Landau's Monto Carlo results for a simulation of the two-dimensional Ising model on a 30×30 lattice.

between the average of the magnetization squared in the vertical columns and horizontal rows and dividing the result by $L^2 = 900$. (This would equal one when the strip formation is perfect and zero when the system is isotropic.) The $m^*(T)$ curve so obtained, for $E = \infty$, is shown in Fig. 7.

Also shown in Fig. 7 is the exact infinite volume equilibrium, $E = 0$, $m^*(T)$ curve as well as some $m^*(T)$ obtained[17] from computer simulations on a 30×30 lattice at $E = 0$. It appears that the nonequilibrium $m^*(T)$ is less steep than the equilibrium one. If this is really the case it would indicate a critical magnetization exponent larger than the equilibrium $1/8$. This would then be similar to what is found for the phase separation of a binary fluid under a shear flow.[18]

The development of two phases at $\beta = \beta_c$ is accompanied at $E = 0$ by a logarithmic singularity in the specific heat as $\beta \to \beta_c$. To find out whether there is a similar behavior in the presence of the field we investigated the specific heat at $E = \infty$ and the saturation current as a function of β. The specific heat, cf. Fig. 8, has a maximum around 0.37 which coincides

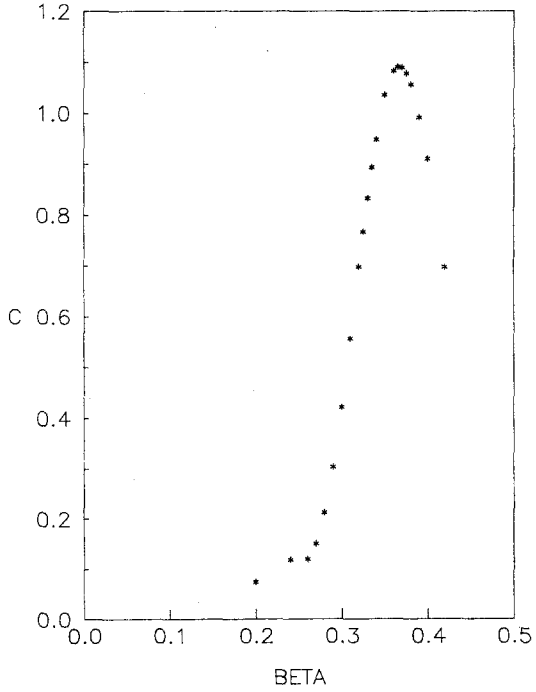


Fig. 8. The specific heat in saturation field. Values were calculated by differentiating a polynomial fit of the energy.

roughly with the critical temperature obtained from the structure function. Compared with the thermal equilibrium, $E = 0$, specific heat, the maximum is not as pronounced. This may also be due to the nature of the singularity in the thermodynamic limit being changed by the field.

We have also looked for any apparent nonsmooth behavior of the current at $\beta_c(E)$. We found that the saturation current seems to have a break around $\beta = 0.3$. In Fig. 9 we plot the saturation current with respect to β. It seems possible that in the thermodynamic limit the slope develops a jump discontinuity.

We also investigated the truncated nearest-neighbor correlations in the vertical and horizontal direction; cf. Fig. 10. Since above $\beta_c(E)$ the column density typically differs from $1/2$, cf. Fig. 6, we truncate the vertical nearest-neighbor correlation in each column separately by the column

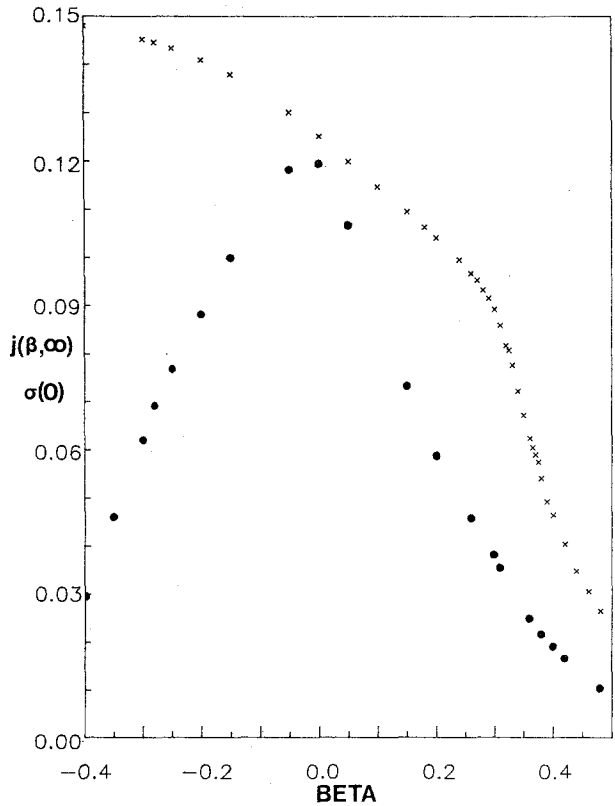


Fig. 9. The crosses represent the saturation current. The solid circles represent the zero field conductivity, $\sigma(0)$.

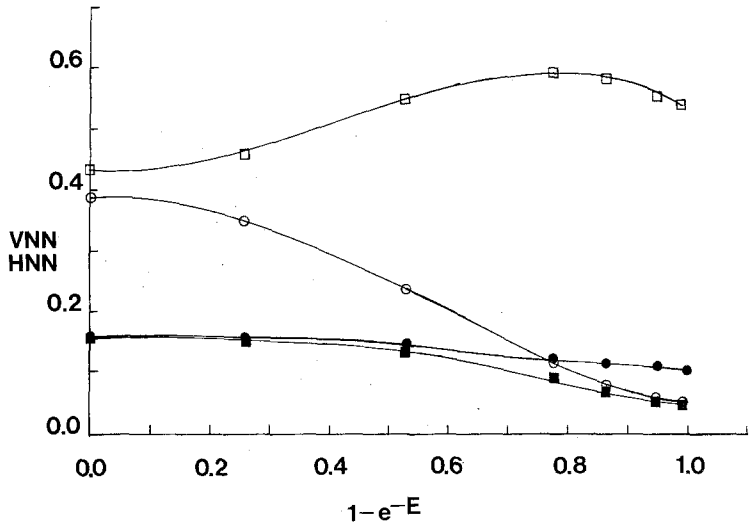


Fig. 10. Truncated vertical nearest-neighbor correlation functions (VNN) and horizontal nearest-neighbor correlation functions (HNN). The open (closed) circles correspond to VNN for $\beta = 0.4$ (0.2). The open (closed) squares correspond to HNN for $\beta = 0.4$ (0.2). The difference between VNN and HNN at $E = 0$ is an indication of statistical fluctuation.

density squared. For consistency we use the same truncation in all other cases too. We see that the correlations become anisotropic for $E \neq 0$. The qualitative behavior of the nearest-neighbor correlations may be explained as resulting from two partially opposing tendencies of the field: (i) to produce strips and thereby to increase correlations and (ii) to decrease the correlations within each column (recall that in one dimension with Metropolis dynamics the $E = \infty$ state is completely random). At small β the ordering tendency dominates and the correlations increase with increasing E. At large β the truncated correlations still increase for small E whereas at the critical field strength, $E \cong 1.5$ for $\beta = 0.4$, strips are formed and the randomization tendency within each column dominates. More study is clearly required for a full understanding of the correlations in this nonequilibrium steady state.

$d = 2$, *Repulsive Interaction.* For $E = 0$ at sufficiently low temperatures the particles try to arrange themselves in a chessboard configuration. For large field the particles jump with rate one to the unoccupied site ahead of them in the $-y$ direction. Therefore the field breaks the antiferromagnetic structure. This mechanism is supported numerically. The average current is larger than the one for the ferromagnetic coupling of the same magnitude; see Ref. 4. The nearest-neighbor correlations increase with

increasing field, i.e., particles become less negatively correlated. The horizontal nearest-neighbor correlation is always less negative than the vertical one. The staggered structure function is only slightly anisotropic. Its maximum, still at either $\mathbf{k} = (2\pi/30, 0)$ or $\mathbf{k} = (0, 2\pi/30)$ is substantially decreased when going from $E = 0$ to $E = \infty$.

6. KUBO'S FORMULA AND BULK DIFFUSIVITY

Kubo's formula relates transport coefficients to the space-time integral over the corresponding current–current correlation function in equilibrium. First we discuss how this is defined for stochastic lattice gases. We then prove the formula for $\sigma(0)$, the zero field conductivity, and show that $\sigma(0)$ is related to the bulk diffusivity for $E = 0$ by an Einstein relation. We then comment on various methods to determine the latter numerically. We close by explaining the corresponding situation for nonzero E.

We define the integrated current through the bond (x, y) by

$$\mathcal{J}(x, y; dt) = \text{number of particles which cross the bond from } x \text{ to } y \text{ minus number of particles which cross the bond from } y \text{ to } x \text{ during the time interval } [t, t + dt]$$

$\mathcal{J}(x, y; dt)$is defined for every "history" (i.e., stochastic trajectory) of the particle system. The density $\mathcal{J}(x, y; t)$ of the integrated current, defined by $\mathcal{J}(x, y; t)\,dt = \mathcal{J}(x, y; dt)$, is the *actual* current through the bond (x, y). It has a δ function with weight 1 at times when a particle jumps from x to y and a δ function with weight -1 at times when a particle jumps from y to x. The actual current should not be confused with the current function (3.1): $j_E(x, y)(\eta)$ is the *average* current *when* the particle configuration is η. For an arbitrary initial probability distribution and an aribitrary dynamics L the average current at time t is given by the average of $\mathcal{J}$ over trajectories followed by an average over configurations. In particular

$$\langle \mathcal{J}(x, y; dt) \rangle = \langle e^{L^* t} j(x, y) \rangle\, dt \tag{6.1}$$

In thermal equilibrium the average current is zero,

$$\langle \mathcal{J}(x, y; dt) \rangle_{0,\rho} = \langle j_0(x, y) \rangle_{0,\rho}\, dt = 0 \tag{6.2}$$

The current–current correlation function in equilibrium is then defined by

$$\langle \mathcal{J}(x, x + e_m; dt)\, \mathcal{J}(y, y + e_n; ds) \rangle \tag{6.3}$$

By translation invariance it depends only on $x - y$ modulo the periodicity of Λ and by time invariance only on $t - s$. In order to compute (6.3) we

replace the integrated current by its short time approximation

$$\mathcal{J}(x, y; [t, t+\tau]) = \eta_{x,t}(1-\eta_{y,t})(1-\eta_{x,t+\tau})\eta_{y,t+\tau} - (1-\eta_{x,t})\eta_{y,t}\eta_{x,t+\tau}(1-\eta_{y,t+\tau}) \tag{6.4}$$

for small τ. Here $\eta_{x,t} = 1$, if site x is occupied at time t, and $\eta_{x,t} = 0$ if site x is empty at time t. We insert the short time approximation in (6.3) with the short time intervals $[0, ds]$ and $[t+ds, t+ds+dt]$. We have to compute then an expectation depending on four different times. At this stage we exploit the Markov property of the stochastic time evolution. Using detailed balance the average equals then

$$-ds\,dt\,\langle j_0(x, x+e_m)e^{L_0^* t} j(y, y+e_n)\rangle \tag{6.5}$$

To obtain the equal time contribution we set in (6.3) both time intervals equal to $[0, \tau]$ and use the short time approximation. For small τ the average equals then

$$\tau\delta_{xy}\delta_{mn}\langle c_0(x, x+e_n, \eta)(\eta_x - \eta_{x+e_n})^2\rangle_{0,\rho} \tag{6.6}$$

Therefore

$$\begin{aligned}\langle \mathcal{J}(x, x+e_m; dt)\,\mathcal{J}(y, y+e_n; ds)\rangle_{0,\rho} &= \delta(t-s)\,dt\,ds\,\delta_{xy}\delta_{mn}\langle c(0, e_n, \eta)(\eta_0 - \eta_{e_n})^2\rangle_{0,\rho} \\ &\quad - dt\,ds\,\langle j_0(x, x+e_m)e^{L_0^* |t-s|} j_0(y, y+e_n)\rangle_{0,\rho}\end{aligned} \tag{6.7}$$

Comparing with (3.5) we obtain for the conductivity

$$\sigma_{mn} = \frac{1}{2}\int_{-\infty}^{\infty}\sum_x \langle \mathcal{J}(x, x+e_m; dt)\,\mathcal{J}(0, e_n; 0)\rangle_{0,\rho} \tag{6.8}$$

The zero-field conductivity equals the space-time integral over the equilibrium current–current correlation function. This is Kubo's famous result derived in another context.[19] The integral over an autocorrelation function is non-negative and therefore $\sigma \geqslant 0$ which is not immediately obvious from (3.5).

In the infinite volume limit the conductivity is still defined by (6.8) with the average now over the infinite volume equilibrium state at inverse temperature β and density ρ. If the equilibrium state has good cluster properties, then the integrand of (6.8) converges in the limit $\Lambda \to \mathbb{Z}^d$, $N \to \infty$, $N/|\Lambda| \to \rho$. However, we have no proof that the finite volume conductivity converges in the same limit to the infinite volume one.

Bulk Diffusion

We proceed now to the connection between the zero-field conductivity and the bulk diffusivity in thermal equilibrium. The bulk diffusivity is

unambiguously defined only for the infinitely extended system. We consider the truncated density–density correlation function

$$\langle \eta_{x,t}\eta_{0,0}\rangle_{0,\rho} - \rho^2 \tag{6.9}$$

in equilibrium at given inverse temperature β and density ρ. One expects that for large x and t the density–density correlation function behaves diffusively as

$$\langle \eta_{x,t}\eta_{0,0}\rangle_{0,\rho} - \rho^2 \cong \chi(\rho) e^{At}(x,0) \tag{6.10}$$

where $e^{At}(q,0)$ is the transition probability for a Brownian particle with diffusion matrix $D(\rho)$. In other words $e^{At}(q,0)$ is the fundamental solution of the diffusion equation

$$\frac{\partial}{\partial t} f(q,t) = \sum_{m,n=1}^{d} \frac{\partial}{\partial q_m} D_{nm}(\rho) \frac{\partial}{\partial q_n} f(q,t) \tag{6.11}$$

By definition $D(\rho)$ is the bulk diffusion coefficient at density ρ. $\chi(\rho)$ is determined by summing both sides of (6.10) over x. Since $\int dq\, e^{At}(q,0) = 1$ by conservation of mass, we obtain

$$\chi(\rho) = \sum_x \left(\langle \eta_x \eta_0 \rangle_{0,\rho} - \rho^2 \right) \tag{6.12}$$

i.e., $\chi(\rho)$ is the static compressibility. By the assumed reflection symmetry $D(\rho)$ is diagonal. Therefore given (6.10) it is reasonable to adopt the following definition of the bulk diffusion coefficient:

$$D_{nn}(\rho) \equiv \lim_{t\to\infty} \frac{1}{2\chi(\rho)} \frac{1}{t} \sum_x (x_n)^2 \left(\langle \eta_{x,t}\eta_{0,0}\rangle_{0,\rho} - \rho^2 \right) \tag{6.13}$$

For high temperature, using the conservation of mass and the good cluster properties of the equilibrium state, we prove in Appendix 2 that the limit (6.13) exists and that

$$D_{mn}(\rho) = \frac{1}{2\chi(\rho)} \int_{-\infty}^{\infty} \sum_{x\in\mathbb{Z}^d} \langle \mathscr{J}(x, x+e_m; dt) \mathscr{J}(0, e_n; 0)\rangle_{0,\rho} \tag{6.14}$$

Comparing with (6.8) we obtain the Einstein relation

$$D_{mn}(\rho) = \frac{1}{\chi(\rho)} \sigma_{mn} \tag{6.15}$$

with σ_{mn} the infinite volume conductivity. By measuring the conductivity one can therefore determine the bulk diffusivity.

There have been various numerical investigations of the bulk diffusivity. We want to briefly compare here the various methods. In principle there are at least five different ways for determining the bulk diffusion constant. In three of them one uses the relationship (6.15). Therefore with

these methods one has to determine the zero-field conductivity and, independently, the static compressibility, which can be done by equilibrium techniques.

(1) One measures the zero field conductivity by measuring the steady-state current for small field. This is the method used here. In practice just a single value of the field suffices. Since the current is an average over all bonds, statistical fluctuations are rather small.

(2) One determines numerically the total current–current correlation function in thermal equilibrium as

$$\frac{1}{|\Lambda|t}\left\langle\left(\sum_{x\in\Lambda}\mathscr{J}(x,x+e_m;[0,t])\right)\left(\sum_{y\in\Lambda}\mathscr{J}(y,y+e_n;[0,t])\right)\right\rangle_{0,N} \tag{6.16}$$

For large times it has to tend to the constant $2\sigma_{mn}$. In this way one evaluates numerically directly the integral (6.14). This method looks rather promising to us. But it has not been employed systematically yet. Sadiq[20] used this method in the case of a two-dimensional system with two conserved quantities near the critical point. The results are modest, in the sense that there are large statistical fluctuations. In our opinion, the reason lies more in the ambitious goal pursued in Ref. 20 than in the inadequacy of the method.

(3) Murch and Thorn[21,22] measured the conductivity for a simple cubic and a two-dimensional honeycomb lattice with both repulsive and attractive interactions. They use a time-dependent method and compute the average drift of an initially sharp mass distribution. The error bars are rather large. It is not clear to us how the precise density dependence of the bulk diffusivity can be accessed by this method.

(4) One prepares the system initially in a slowly, sinusoidally varying density. This density decays then to uniform equilibrium and the rate of decay is a measure of the bulk diffusion coefficient. Kutner, Binder, and Kehr[23] used this method in the case of a three-dimensional system on a simple cubic lattice with attractive interactions. They cover the whole density range at various temperatures. To our knowledge, this is the most systematic measurement of a bulk diffusivity. It is not so clear to us how well this method can handle statistical errors and finite wavelength and frequency effects.

(5) One establishes the system in a nonequilibrium steady state by imposing different densities at the boundaries. This is achieved by creation and destruction of particles at the boundary. In the steady state the mass current is proportional to the bulk diffusivity for small density difference. Murch[24] used this method for a three-dimensional simple cubic lattice with attractive and repulsive interactions. Since the current is an average over all bonds, statistical errors are presumably small. However, finite size

effects and nonlinear dependence on the density difference may be important.

Certainly, all five methods work in principle: which one of the methods gives the must reliable numerical value for the bulk diffusivity still waits for clarification. In our opinion, stationary methods are closest to the theoretical definition.

So far we have focused on the zero-field conductivity and diffusivity. The finite field diffusivity is still given by the space-time integral over the current–current correlation function. Since to our knowledge this has not been given in the literature, we explain the formal derivation in Appendix 3. There cannot be any simple relationship like (6.15) between diffusivity and conductivity at nonzero field. To see this we define the rates

$$\tilde{c}_E(x, y, \eta) = \alpha(E) c_E(x, y, \eta) \tag{6.17}$$

with $\alpha(E) > 0$ and independent of x, y, and η and c_E satisfying the detailed balance condition (1.5). $\tilde{c}_E$ then also satisfies the detailed balance condition. Since (6.17) just amounts to a change of the time scale, the stationary states of the dynamics with rates $\tilde{c}_E$ are the same as those with c_E. Therefore

$$\tilde{j}_n(\beta, E) \equiv \langle \tilde{j}_n(0, e_n) \rangle_{E,\rho} = \alpha(E) j_n(\beta, E) \tag{6.18}$$

and

$$\tilde{\sigma}_{mn}(\beta, E) = \left[\frac{\partial}{\partial E_n} \alpha(E) \right] j_m(\beta, E) + \alpha(E) \sigma_{mn}(\beta, E) \tag{6.19}$$

By an appropriate choice of $\alpha(E)$ we can produce essentially any current–field characteristic we want. In particular, the finite field conductivity can be negative, which is not possible for the bulk diffusivity.

Of course, we can still follow the computation in Appendix 1 and express the conductivity as

$$\sigma_{mn}(E) = \left\langle \frac{\partial}{\partial E_n} j_E(0, e_m) \right\rangle_{E,\rho} + \int_0^\infty dt \sum_\eta \rho_{E,N}(\eta) \left(\frac{\partial}{\partial E_n} L_E^* \right) e^{L_E^* t} j_E(0, e_m)(\eta) \tag{6.20}$$

7. ENTROPY PRODUCTION

In linear irreversible thermodynamics the steady state can be obtained as the minimum of the entropy production.[25] We take this as our motivation to investigate here whether a corresponding principle could be

valid on the level of stochastic particle dynamics. We comment at the end of this section on the entropy production for a classical particle system in contact with heat reservoirs.

We define the entropy production $\sigma(\rho)$ in the state ρ as

$$\sigma(\rho) = \text{change in system entropy} + \text{flow of entropy from the system to the surroundings}$$

The change in the system entropy in the state ρ is

$$-\frac{d}{dt}\sum_{\eta}\rho_t(\eta)\log\rho_t(\eta)\Big|_{\rho_t=\rho} \tag{7.1}$$

with ρ_t governed by the master equation (1.2). The entropy flow to the surroundings has two contributions: (a) there is a change in the energy of the system when a transition changes the number of nearest neighbors. The corresponding entropy flow is

$$-\beta\sum_{\eta}\rho(\eta)L^*H(\eta) \tag{7.2}$$

(b) There is work done by the electric field which is absorbed by the surroundings as heat at the temperature β^{-1}. This is equal to E times the average current

$$-\tfrac{1}{2}\sum_{\eta}\rho(\eta)\sum_{\substack{x,y\\|x-y|=1}}c_E(x,y,\eta)E\cdot(x-y)(\eta_x-\eta_y) \tag{7.3}$$

Remember that E corresponds to βE. Therefore, adding (7.1) to (7.3), we obtain

$$\begin{aligned}\sigma(\rho) = -\tfrac{1}{2}\sum_{\substack{x,y\\|x-y|=1}}\sum_{\eta}\{&\left[c_E(x,y,\eta^{xy})\rho(\eta^{xy})-c_E(x,y,\eta)\rho(\eta)\right]\log\rho(\eta)\\&+\rho(\eta)c_E(x,y,\eta)\beta\left(H(\eta^{xy})-H(\eta)\right)\\&+\rho(\eta)c_E(x,y,\eta)E\cdot(x-y)(\eta_x-\eta_y)\}\end{aligned} \tag{7.4}$$

Note that the first two terms are the time derivative of the "free energy," $E - TS$, whereas the third term cannot be written as a time derivative.

Let $\nu(\rho) = e^{\beta H(\eta)}\rho(\eta)$. Then using the detailed balance condition (1.5) we can rewrite (7.4) as

$$\begin{aligned}\sigma(\nu e^{-\beta H}) = \tfrac{1}{4}\sum_{\substack{x,y\\|x-y|=1}}\sum_{\eta}&c_E(x,y,\eta)e^{-\beta H(\eta)}\left[\nu(\eta)-e^{E\cdot(x-y)(\eta_x-\eta_y)}\nu(\eta^{xy})\right]\\&\times\left[\log\nu(\eta)-\log e^{E\cdot(x-y)(\eta_x-\eta_y)}\nu(\eta^{xy})\right]\end{aligned} \tag{7.5}$$

The function $f(x, y) = (x - y)(\log x - \log y)$ is non-negative for $x, y > 0$ and we conclude that

$$\sigma(\rho) \geqslant 0 \tag{7.6}$$

In fact, for $E \neq 0$, $\sigma(\rho) > 0$ because $\sigma(\rho) = 0$ if and only if $\nu(\eta) = e^{E\cdot(x-y)(\eta_x - \eta_y)}\nu(\eta^{xy})$, which cannot be satisfied because of the periodic boundary conditions. Since for $0 \leqslant \lambda \leqslant 1$, $f((1-\lambda)x_1, (1-\lambda)y_1) + f(\lambda x_2, \lambda y_2) \leqslant (1-\lambda)f(x_1, y_1) + \lambda f(x_2, y_2)$ with strict inequality if $(x_1, y_1) \neq \alpha(x_2, y_2)$, σ is *strictly* convex, i.e.,

$$\sigma\big((1-\lambda)\rho_1 + \lambda\rho_2\big) < (1-\lambda)\sigma(\rho_1) + \lambda\sigma(\rho_2) \tag{7.7}$$

for $\rho_1 \neq \rho_2$. Therefore σ has a unique minimum $\rho_{E,\min}$, the state of minimal entropy production at given field E.

How is $\rho_{E,\min}$ linked to the steady state $\rho_{E,N}$ defined by (2.1)? From simple examples we learn that $\rho_{E,\min} \neq \rho_{E,N}$ in general. However to first order in E they agree, i.e.,

$$\frac{\partial}{\partial E_n}\rho_{E,N}\bigg|_{E=0} = \frac{\partial}{\partial E_n}\rho_{E,\min}\bigg|_{E=0} \tag{7.8}$$

We computed already the left-hand side of (7.8), see (3.3)–(3.5), as

$$-\left(\int_0^\infty dt \sum_x e^{L_0^\dagger t} j_0(x, x + e_n)\right) Z^{-1} e^{-\beta H} \tag{7.9}$$

To obtain the right-hand side we take the variational derivative of σ at $\nu_{E,\min}$. This gives

$$\sum_{\substack{x,y \\ |x-y|=1}} c_E(x, y, \eta)\big[1 - e^{E\cdot(x-y)(\eta_x - \eta_y)}\nu_{E,\min}(\eta^{xy})\nu_{E,\min}(\eta)^{-1} + \log\nu_{E,\min}(\eta) - \log\nu_{E,\min}(\eta^{xy}) - E\cdot(x-y)(\eta_x - \eta_y)\big] = 0 \tag{7.10}$$

We solve (7.10) to first order in E and obtain

$$\sum_{\substack{x,y \\ |x-y|=1}} c_0(x, y, \eta)\left\{\frac{\partial}{\partial E_n}\big[\nu_{E,\min}(\eta^{xy}) - \nu_{E,\min}(\eta)\big]\right\}\bigg|_{E=0} = \sum_x c_0(x, x + e_n, \eta)(\eta_x - \eta_{x+e_n}) \tag{7.11}$$

in agreement with (7.8) and (7.9).

We remark that the agreement between steady state and state of minimal entropy production to linear order in E relies in an essential way on the stochastic nature of the dynamics. If we consider a system of

classical particles in contact with stochastic heat reservoirs[26] then even to linear order in the temperature difference the steady state does not agree with the state of minimal entropy production. The reason is that, since the free energy is invariant under the Hamiltonian time evolution, the entropy production for a given measure is independent of the Hamiltonian. On the other hand the first-order correction to the steady state depends on the particular form of the Hamiltonian.

8. THE INFINITE-VOLUME LIMIT

Physically, one is typically interested in the bulk properties of the system. Therfore, as in thermal equilibrium, surface effects should be eliminated by taking the thermodynamic limit where the size of the system tends to infinity keeping the density fixed. We choose then a sequence Λ_j of periodic boxes, $\Lambda_j \nearrow \mathbb{Z}^d$ as $j \to \infty$, and a sequence N_j of number of particles such that $\lim_{j\to\infty} N_j/|\Lambda_j| = \rho$ and study the limit of the correlation functions

$$\lim_{j\to\infty} \left\langle \prod_{x\in A} \eta_x \right\rangle_{E,N_j/|\Lambda_j|} \equiv \int \mu_{E,\rho}(d\eta) \prod_{x\in A} \eta_x \tag{8.1}$$

where A is an arbitrary finite set of sites. By compactness the limit taken along suitable subsequences exists and defines the infinite volume state $\mu_{E,\rho}$. By construction $\mu_{E,\rho}$ is stationary and translation invariant.

By just using compactness one loses any specific information about the infinite volume states. But we conjecture that they have the following properties: For sufficiently small β for any density ρ and field E the limit state $\mu_{E,\rho}$ should cluster in the sense that truncated correlation functions decay at infinity. On the other hand for given E and $\beta > \beta_c(E)$ one should have two density ranges $0 \leqslant \rho \leqslant \rho_g$ and $\rho_l \leqslant \rho \leqslant 1$ such that for every ρ within that range $\mu_{E,\rho}$ has good clustering whereas for $\rho_g < \rho < \rho_l$ the limit (8.1) will be a suitable superposition of the states μ_{E,ρ_g} and μ_{E,ρ_l}. In particular the limit correlation functions in (8.1) do not cluster and the system exhibits long-range order. As already noted the segregation curve has to be symmetric around $\rho = 1/2$. A typical configuration for μ_{E,ρ_l} would have an infinite cluster of particles with holes stretched along the field direction. If in this configuration we interchange particles and empty sites, then we obtain a typical configuration for μ_{E,ρ_g}.

Another approach to the infinite volume limit is to study directly the stationary states of the infinite volume dynamics. We consider the space Ω of all configurations on $\mathbb{Z}^d$, i.e., $\Omega = \{0,1\}^{\mathbb{Z}^d}$. It has been shown[25] that the evoluation equation

$$\frac{d}{dt} f_t(\eta) = (L_E^* f_t)(\eta) \tag{8.2}$$

with

$$(L_E^* f)(\eta) = \frac{1}{2} \sum_{x,y \in \mathbb{Z}^d} c_E(x, y, \eta)\big[f(\eta^{xy}) - f(\eta)\big] \tag{8.3}$$

acting on local functions has a unique solution $e^{L_E^* t}$ on $C(\Omega)$, the space of bounded and continuous functions on Ω. The kernel $e^{L_E^* t}(\eta \mid d\eta')$ defines the transition probability for the infinite system. Invariant probability measures are characterized by

$$\int_\Omega \mu(d\eta)(L_E^* f)(\eta) = 0$$

for all local functions f. The problem is then to classify all invariant states for given parameters E and β. One class of stationary states is provided by the construction (8.1). These steady states are translation invariant and carry a current. The dynamics in these states is not reversible. Another class of invariant states is obtained by the infinite volume of the Gibbs state (1.6). Their density tends to unity in the direction of the field and to zero opposite to the direction of the field. In these states the dynamics is reversible. If we restrict out attention to steady states which are translation invariant, then we would hope that they coincide exactly with the ones obtained through the limit (8.1).

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APPENDIX 1

We fill in the details in deriving (3.5). Differentiating the detailed balance condition (1.5) at $E_n = 0$ one obtains

$$\begin{aligned} c_0'(x, x + e_m, \eta) &= c_0'(x, x + e_m, \eta^{x\,x+e_m}) e^{-\beta(H(\eta^{x\,x+e_m}) - H(\eta))} \\ &\quad + c_0(x, x + e_m, \eta) e^{-\beta(H(\eta^{x\,x+e_m}) - H(\eta))}(\eta_x - \eta_{x+e_m})\delta_{mn} \end{aligned} \tag{A.1}$$

Inserting (A.1) in the first term of (3.4) yields

$$\tfrac{1}{2}\langle c(x, x + e_n)(\eta_x - \eta_{x+e_n})^2 \rangle \delta_{mn} \tag{A.2}$$

Differentiating the invariance condition at $E_n = 0$ one obtains

$$L\rho_{0,N}' + L'\rho_{0,N} = 0 \tag{A.3}$$

By (A.1)

$$L'\rho_{0,N} = -\left[\sum_x j(x, x + e_m)\right]\rho_{0,N} \tag{A.4}$$

Using again detailed balance the first-order correction is then

$$\rho'_{0,N} = -\left[\int_0^\infty dt \sum_x e^{L^* t} j(x, x + e_m)\right]\rho_{0,N} \tag{A.5}$$

Inserting in the second term of (3.4) gives the desired result.

APPENDIX 2

We prove the existence of the limit (6.13) under the condition of sufficiently high temperatures. Our argument does not exclude the physically absurd possibility $D(\rho) \equiv 0$. In the following manipulations we invoke the following lemma.

Lemma. Let f and g be local functions and let τ_x denote the shift by $x \in \mathbb{Z}^d$. If ρ and β satisfy the high-temperature condition $|\beta\mu(\rho)| + 2d|\beta| < \pi/4$ with $\mu(\rho)$ the chemical potential as a function of ρ, then

$$|\langle g\tau_x e^{L^* t} f\rangle_{\rho,\beta}| \leqslant c_1 e^{-c_2|x|} \tag{A.6}$$

with positive constants c_1, c_2 depending on g, f, and t. Here $\langle \cdot \rangle_{\rho,\beta}$ denotes the average over the infinite volume equilibrium state at inverse temperature β and density ρ.

Proof. Since f is local, the existence of the dynamics implies that $e^{L^* t} f$ is quasilocal in the sense that, for fixed t, it depends only exponentially little on the occupation variables far out. The explicit dependence follows from estimates similar to those of Holley and Stroock.[28,29] Our high-temperature condition guarantees exponential clustering.[29,30] Both results together imply (A.6). ■

The constant c_1 grows exponentially with t. (A.6) is therefore useful only for finite t.

From the definition of the integrated current we deduce the conservation law

$$\eta_{x,t} - \eta_{x,0} = \sum_{m=1}^{d} \{\mathscr{J}(x - e_m, x; [0,t]) - \mathscr{J}(x, x + e_m; [0,t])\} \tag{A.7}$$

for every history. Because of the bound (A.6) the following identities are

valid:

$$\frac{1}{t}\sum_x (x_n)^2\big(\langle \eta_{x,t}\eta_{0,0}\rangle_{\rho,\beta} - \rho^2\big)$$

$$= -\frac{1}{2t}\sum_x (x_n)^2\langle(\eta_{x,t} - \eta_{x,0})(\eta_{0,t} - \eta_{0,0})\rangle_{\rho,\beta} + \frac{1}{t}\sum_x (x_n)^2\big(\langle \eta_x\eta_0\rangle_{\rho,\beta} - \rho^2\big)$$

$$= \frac{1}{t}\sum_x \langle \mathscr{J}(x, x+e_n; [0,t])\mathscr{J}(0, e_n; [0,t])\rangle_{\rho,\beta}$$

$$+ \frac{1}{t}\sum_x \big(\langle \eta_x\eta_0\rangle_{\rho,\beta} - \rho^2\big) \tag{A.8}$$

We used here detailed balance and partially integrated twice. As $t\to\infty$ the second term vanishes. For the first term we use (6.7). Then

$$(\text{A.8}) = \langle c(0, e_n, \eta)(\eta_0 - \eta_{e_n})^2\rangle_{\rho,\beta}$$

$$- \frac{1}{t}\int_0^t ds \int_0^t ds'\, h(|s - s'|) + O(1/t) \tag{A.9}$$

with

$$h(t) = \sum_x (x, x+e_n) e^{L_0^\dagger |t|} j(0, e_n)\rangle_{0,\rho}$$

The integrand of the second term is positive. Therefore its limit as $t\to\infty$ exists and is either finite or infinite. If it would be infinite, then the sum of the first two terms would be negative, which contradicts the positivity of the total current–current correlation function. We conclude then that the limit (6.13) exists and is given by (6.14).

The argument given yields no information about the decay of $h(t)$. In principle the two terms of (3.5) could cancel each other in the limit $t\to\infty$.

APPENDIX 3

We want to show here that the bulk diffusivity at finite E is still given by the space-time integral over the truncated current–current correlation function. We have too little control over the dynamics and over the stationary state to make our argument rigorous. We assume the existence of an infinite volume state $\langle\cdot\rangle_\rho$ with density ρ which is stationary in time, invariant under spatial translation, and has some reasonable clustering in space and time. The detailed balance condition (1.5) is not used. In the case of a constant electric field $\langle\ \rangle_\rho \equiv \langle\ \rangle_{E,\rho}$.

The basic assumption is that the truncated density–density correlation function,

$$\langle \eta_{x,t}\eta_{0,0}\rangle_\rho - \rho^2 \tag{A.10}$$

behaves for large x and t as the fundamental solution of the linearized macroscopic equation

$$\frac{\partial}{\partial t}\rho(q,t) = -\sum_{m=1}^{d}\left(\frac{\partial}{\partial \rho} j_m(\rho)\right)\frac{\partial}{\partial q_m}\rho(q,t) + \sum_{m,n=1}^{d}\frac{\partial}{\partial q_m} D_{mn}(\rho)\frac{\partial}{\partial q_n}\rho(q,t) \tag{A.11}$$

Here, by definition, $D(\rho)$ is the bulk diffusion matrix and $j(\rho)$ is the stationary current, i.e., $j_m(\rho) = \langle j(0,e_m)\rangle_\rho$. This allows us to identify the macroscopic quantities with microscopic expressions. The constant drift term appears because we no longer assume reversibility.

The first moment of (A.10) should grow linearly in t and identify then the correct drift. The normalization is

$$\chi(\rho) = \sum_x \left(\langle \eta_x\eta_0\rangle_\rho - \rho^2\right) \tag{A.12}$$

Using the conservation law we obtain

$$\begin{aligned}
&\frac{1}{\chi(\rho)}\sum_x x_m\left(\langle \eta_{x,t}\eta_{0,0}\rangle_\rho - \rho^2\right)\\
&\quad = \frac{1}{\chi(\rho)}\sum_x x_m\left(\langle (\eta_{x,t}-\eta_{x,0})\eta_{0,0}\rangle_\rho + \langle \eta_x\eta_0\rangle_\rho - \rho^2\right)\\
&\quad = \frac{1}{\chi(\rho)}\sum_x x_m\left(\sum_{n=1}^{d}\langle \left(\mathscr{J}(x-e_n,x;[0,t]) - \mathscr{J}(x,x+e_n;[0,t])\right)\eta_{0,0}\rangle_\rho\right)\\
&\quad = \frac{1}{\chi(\rho)}\int_0^t ds\sum_x\left[\langle \eta_x e^{L^* s} j(0,e_m)\rangle_\rho - \langle \eta_0\rangle_\rho\langle j(0,e_m)\rangle_\rho\right]\\
&\quad = t\frac{1}{\chi(\rho)}\sum_x\left(\langle \eta_x j(0,e_m)\rangle_\rho - \langle \eta_0\rangle_\rho\langle j(0,e_m)\rangle_\rho\right)
\end{aligned} \tag{A.13}$$

Let

$$\langle\cdot\rangle_{\rho(\lambda)} = Z(\lambda)^{-1}\left\langle \cdot \exp\left(\lambda\sum_x \eta_x\right)\right\rangle_\rho \tag{A.14}$$

Then

$$\sum_x (\langle \eta_x j(0,e_m)\rangle_\rho - \langle \eta_0\rangle_\rho \langle j(0,e_m)\rangle_\rho)$$

$$= \frac{\partial}{\partial\lambda}\langle j(0,e_m)\rangle_{\rho(\lambda)}\Big|_{\lambda=0} = \left[\frac{\partial}{\partial\rho} j_m(\rho)\right]\frac{\partial\rho(\lambda)}{\partial\lambda}\Big|_{\lambda=0} = \chi(\rho)\frac{\partial}{\partial\rho} j_m(\rho) \tag{A.15}$$

which shows that

$$\frac{1}{\chi(\rho)}\sum_x x_m\big(\langle \eta_{x,t}\eta_{0,0}\rangle_\rho - \rho^2\big) = t\frac{\partial}{\partial\rho} j_m(\rho)$$

as claimed above.

To identify the bulk diffusion matrix we define

$$\sum_{m,n=1}^{d} a_m^* a_n D_{mn}(\rho)$$

$$= \lim_{t\to\infty}\frac{1}{2t}\left[\frac{1}{\chi(\rho)}\sum_x\left|\sum_{m=1}^{d} a_m x_m\right|^2\big(\langle \eta_{x,t}\eta_{0,0}\rangle_\rho - \rho^2\big)\right.$$

$$\left. - \left|\frac{1}{\chi(\rho)}\sum_x\sum_{m=1}^{d} a_m x_m\big(\langle \eta_{x,t}\eta_{0,0}\rangle_\rho - \rho^2\big)\right|^2\right] \tag{A.16}$$

We follow the same steps as in (A.8) and obtain

$$\frac{1}{2t}\frac{1}{\chi(\rho)}\sum_x\left|\sum_{m=1}^{d} a_m x_m\right|^2\big(\langle \eta_{x,t}\eta_{0,0}\rangle_\rho - \rho^2\big)$$

$$= \sum_x\sum_{m,n=1}^{d} a_m^* a_n(\langle \mathscr{J}(x,x+e_m;[0,t])\mathscr{J}(0,e_n;[0,t])\rangle_\rho$$

$$- t\langle j(0,e_m)\rangle_\rho t\langle j(0,e_n)\rangle_\rho) \tag{A.17}$$

The correct spatial truncation appears through the partial integration. To compute the current–current correlation function we use again the short-time approximation (6.4) and follow the argument given there. Then

$$\langle \mathscr{J}(0,e_m;ds)\mathscr{J}(x,x+e_n;dt)\rangle_\rho$$

$$= \delta(t-s)\,dt\,ds\,\delta_{mn}\delta_{x0}\langle c(0,e_n)(\eta_0 - \eta_{e_n})^2\rangle_\rho$$

$$+ dt\,ds\,\langle j(0,e_m)\partial_{0e_m} e^{L^*|t-s|} j(x,x+e_n)\rangle_\rho \tag{A.18}$$

Here ∂_{xy} interchanges the occupations at x and y, i.e., $(\partial_{xy}f)(\eta) = f(\eta^{xy})$.

Note that in thermal equilibrium detailed balance implies $\partial^*_{0e_m} j(0, e_m) = -j(0, e_m)$ with the definition $\langle \partial^*_{0e_m} j(0, e_m) f \rangle_\rho = \langle j(0, e_m) \partial_{0e_m} f \rangle_\rho$ for all local functions f. Inserting (A.18) in (A.17) we obtain

$$\sum_{m,n=1}^{d} a_m^* a_n D_{mn}(\rho)$$

$$= \frac{1}{2\chi(\rho)} \sum_{n=1}^{d} |a_n|^2 \langle c(0, e_n)(\eta_0 - \eta_{e_n})^2 \rangle_\rho$$

$$+ \sum_{m,n=1}^{d} a_m^* a_n \left\{ \lim_{t\to\infty} \frac{1}{2t} \int_0^t ds \int_0^t ds' \right.$$

$$\times \left[\frac{1}{\chi(\rho)} \sum_x \left(\langle j(0, e_m) \partial_{0e_m} e^{L^*|s-s'|} j(x, x + e_n) \rangle_\rho \right.\right.$$

$$\left. - \langle j(0, e_m) \rangle_\rho \langle j(0, e_n) \rangle_\rho \right)$$

$$\left.\left. - \left(\frac{\partial}{\partial \rho} j_m(\rho) \right) \left(\frac{\partial}{\partial \rho} j_n(\rho) \right) \right] \right\} \tag{A.19}$$

We have to check that (A.19) contains the correct truncation in time. For this we have to study

$$\lim_{t\to\infty} \frac{1}{\chi(\rho)} \sum_x \left(\langle \tau_x g e^{L^* t} f \rangle_\rho - \langle g \rangle_\rho \langle f \rangle_\rho \right) \tag{A.20}$$

where $g = \partial^*_{0e_m} j(0, e_m)$ and $f = j(0, e_n)$. We write (A.20) as

$$\lim_{t\to\infty} \frac{1}{\chi(\rho)} \frac{\partial}{\partial \lambda} Z(\lambda)^{-1} \left\langle (E^{L^* t} f) \exp\left(\lambda \sum_x \tau_x g \right) \right\rangle_\rho \Big|_{\lambda=0}$$

and interchange the differentiation and the limit. Then

(A.20)

$$= \frac{\partial}{\partial \lambda} \lim_{t\to\infty} Z(\lambda)^{-1} \left\langle (e^{L^* t} f) \exp\left(\lambda \sum_x \tau_x g \right) \right\rangle_\rho \Big|_{\lambda=0} \frac{\partial}{\partial \lambda} \langle f \rangle_{\rho(\lambda)} \Big|_{\lambda=0} \tag{A.21}$$

where $\langle \cdot \rangle_{\rho(\lambda)}$ is the stationary state with density $\rho(\lambda)$ obtained in the limit

as $t \to \infty$ of $Z(\lambda)^{-1}\langle \cdot \exp(\lambda \sum_x \tau_x g)\rangle_\rho$. Therefore

$$(A.20) = \frac{1}{\chi(\rho)}\left(\frac{\partial}{\partial \rho}\langle f\rangle_\rho\right)\frac{\partial \rho(\lambda)}{\partial \lambda}\bigg|_{\lambda=0}$$

$$= \left[\frac{1}{\chi(\rho)}\sum_x (\langle \eta_x f\rangle_\rho - \langle \eta_0\rangle_\rho \langle f\rangle_\rho)\right]\left[\frac{1}{\chi(\rho)}\sum_x (\langle \eta_x g\rangle_\rho - \langle \eta_0\rangle_\rho \langle g\rangle_\rho)\right] \tag{A.22}$$

where the same argument as in (A.15) is used. Inserting $g = \partial^*_{0e_m} j(0, e_m)$ and $f = j(0, e_n)$ and using (A.15) and

$$\sum_x (\langle \eta_x \partial^*_{0e_m} j(0, e_m)\rangle_\rho - \langle \eta_0\rangle_\rho \langle j(0, e_m)\rangle_\rho)$$

$$= \sum_x (\langle j(0, e_m)\partial_{0e_m}\eta_x\rangle_\rho - \langle \eta_0\rangle_\rho \langle j(0, e_m)\rangle_\rho)$$

$$= \sum_x (\langle \eta_x j(0, e_m)\rangle_\rho - \langle \eta_0\rangle_\rho \langle j(0, e_m)\rangle_\rho) \tag{A.23}$$

we find that the truncation is indeed as given in (A.19).

Assuming the integrable decay of the total current correlation function

$$D_{mn}(\rho) = \frac{1}{2\chi(\rho)}\langle c(0, e_n)(\eta_0 - \eta_{e_n})^2\rangle_\rho \delta_{mn}$$

$$+ \int_{-\infty}^{\infty} dt \left\{ \frac{1}{\chi(\rho)}\sum_x \left[\langle j(0, e_m)\partial_{0e_m} e^{L^*|t|} j(x, x + e_n)\rangle_\rho \right.\right.$$

$$\left. - \langle j(0, e_m)\rangle_\rho \langle j(0, e_n)\rangle_\rho\right]$$

$$\left. - \left[\frac{\partial}{\partial \rho} j_m(\rho)\right]\left[\frac{\partial}{\partial \rho} j_n(\rho)\right]\right\} \tag{A.24}$$

Note that the spatial sum and the time integration cannot be interchanged. First one has to perform the spatial truncation and the spatial sum. The total current–current correlation function has then to be truncated in time and integrated over all times. The same structure was found for the one-dimensional system of hard rods.[31]

APPENDIX 4

We want the steady state to be independent of E. (2.1) results then in

$$\sum_{\substack{x,y \\ |x-y|=1}} c_E(x, y, \eta)(e^{E\cdot(x-y)(\eta_x - \eta_y)} - 1) = 0 \tag{A.25}$$

which should be read as an equation for the jump rates. To solve it in one dimension we use that the rates depend only on the nearest neighbors of the bond $(x, x+1)$ and set

$$\begin{aligned} c_E(x, x+1; 1, 1, 0, 1) &= \alpha_1(E) \\ c_E(x, x+1; 0, 1, 0, 1) &= \alpha_2(E) \\ c_E(x, x+1; 1, 1, 0, 0) &= \alpha_3(E) \\ c_E(x, x+1; 0, 1, 0, 0) &= \alpha_4(E) \end{aligned} \tag{A.26}$$

with the rates in the opposite direction determined by the detailed balance condition (1.5). (A.25) is then satisfied only if the rates are related by

$$\begin{aligned} \alpha_2 &= e^{\beta}\alpha_3 \\ \alpha_1 - \alpha_2 - \alpha_3 + \alpha_4 &= 0 \end{aligned} \tag{A.27}$$

In linear response this result was noted in Ref. 13. If we require in addition to (A.27) that for given β (1.7) should be satisfied for all E with a function ϕ independent of E, then this is possible only for $\beta = 0$ and $\phi(h) = e^{-h/2}$.

In more than one dimension (A.25) has as only solution $c_E(x, y, \eta) = e^{-E\cdot(x-y)(\eta_x - \eta_y)/2}$.

REFERENCES

1. Cf. J. L. Lebowitz, *Prog. Theor. Phys.* **64**:35 (1978); and references therein.
2. W. Dietrich, P. Fulde, and I. Peschel, *Adv. Phys.* **29**:527 (1980); and references therein.
3. L. Onsager, *Phys. Rev.* **65**:117 (1944); *Nuovo Cimento Suppl.* **6**:261 (1949).
4. S. Katz, J. L. Lebowitz, and H. Spohn, *Phys. Rev. B* **28**:1655 (1983).
5. A. D. LeClaire, in *Physical Chemistry. An Advanced Treatise*, Vol. 10, p. 261, edited by H. Eyring, D. Henderson and W. Jost (Academic Press, New York, 1970).
6. C. Kipnis and S. R. S. Varadhan, to appear.
7. N. Metropolis, A. W. Rosenbluth, M. M. Rosenbluth, A. H. Teller, and E. Teller, *J. Chem. Phys.* **21**:1087 (1953).
8. Cf. *Monte Carlo Methods in Statistical Mechanics*, K. Binder, ed. (Springer, Berlin, 1979).
9. J. C. Kimball and L. W. Adams, *Phys. Rev. B* **18**:5851 (1978).
10. B. I. Halperin, *Phys. Rev. B* **8**:4437 (1973).
11. H. Mori, *Prog. Theor. Phys.* **33**:423 (1965).
12. H. Singer, Diploma thesis, Universität, Berlin, 1980, unpublished.
13. H. Singer and I. Peschel, *Z. Phys.* **B39**:333 (1980).
14. R. Zeyher, *Solid State Commun.* **36**:33 (1980).
15. M. F. Sykes and M. E. Fisher, *Physica* **28**:919 (1962).
16. C. Domb and M. F. Sykes, *Proc. R. Soc. (London)* **A235**:247 (1956).
17. D. P. Landau, *Phys. Rev. B* **13**:2997 (1976).
18. A. Onuki and K. Kawasaki, *Ann. Phys. (N.Y.)* **121**:456 (1979); and **131**:217 (1981). D. Beysens and M. Gbadamassi, *Phys. Rev. A* **22**:2250 (1980).
19. R. Kubo, *J. Phys. Soc. Jpn.* **12**:570 (1957).
20. A. Sadiq, *Phys. Rev. B* **9**:2299 (1974).
21. G. E. Murch and R. Thorn, *J. Phys. Chem. Solids* **39**:1301 (1978).

22. G. E. Murch and R. Thorn, *Phil. Mag.* **37**:85 (1977).
23. R. Kutner, K. Binder, and K. W. Kehr, Diffusion in concentrated lattice gases II, preprint, KFA Jülich.
24. G. E. Murch, *Phil. Mag.* **A41**:159 (1980).
25. Cf. S. R. de Groot and P. Mazur, *Non Equilibrium Thermodynamics* (North-Holland, Amsterdam, 1982).
26. J. L. Lebowitz and P. G. Bergmann, *Phys. Rev.* **99**:578 (1955); *Ann. Phys. (N.Y.)* **1**:1 (1957).
27. T. M. Liggett, The stochastic evolution of infinite systems of interacting particles, *Lecture Notes Math.* **598**:249 (1977).
28. R. A. Holley and D. W. Strook, *Commun. Math. Phys.* **48**:249 (1976).
29. R. A. Holley and D. W. Strook, *Z. Wahrscheinlichkeitstheorie verw. Gebiete* **35**:87 (1976).
30. R. B. Israel, *Commun. Math. Phys.* **50**:245 (1976).
31. H. Spohn, *Ann. Phys. (N.Y.)* **141**:353 (1982).