Steady-State Properties of a Finite System Driven by a Chemical-Potential Gradient

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A two-dimensional lattice-gas model with repulsive interactions periodically infinite in one dimension and finite in the other is driven into a mass-transporting steady state by asymmetric chemical potentials applied at the open edges. By computer-simulation techniques the steady-state current, the mass profile, and the order-parameter profile are calculated. The driven system is found to display a nonequilibrium phase behavior controlled by the mass flux.

A new dimension has been added to the field of phase transitions and critical phenomena by the study of steady-state nonequilibrium systems.[1-9] Only little is known about such systems, in contrast to equilibrium phase-transition phenomena. Prototypical steady-state nonequilibrium systems are driven diffusive systems which are not described by a global Hamiltonian. These systems are thermodynamically open and continuously receive energy from external sources. A fundamental question concerns the nature of phase transitions in such systems and their possible universal features. Our main knowledge about driven diffusive systems derives from studies of a particular microscopic lattice-gas model proposed by Katz, Lebowitz, and Spohn[1,2] as a model of ion transport in fast ionic conductors driven by external homogeneous electric fields. This model has been investigated by field-theoretical methods[4,7] and by computer-simulation techniques,[1,2,7,8] in the cases of both attractive and repulsive interactions. It has been found that the driven steady-state system undergoes a nonequilibrium phase transition. In the case of attractive interactions (conserved order parameter),[1,2] the transition temperature increases with the field strength and the critical exponents deviate from their equilibrium values. In the case of repulsive interactions (nonconserved order parameter),[7] the transition temperature decreases with field strength and there are indications of a tricritical point. In this case the critical exponents remain at their equilibrium values along the line of continuous transitions. Another interesting aspect of these systems concerns the properties of the interfaces between phase-separated regions which occur at low temperatures in the driven system with attractive interactions.[8,10,11] The characteristic feature of all these studies is that they refer to systems driven by a long-range field which acts with equal strength at each particle throughout the system. Hence the effects found persist in the thermodynamic limit.

Under many physical circumstances, however, external driving forces and symmetry-breaking fields only act directly on a finite portion of the system, e.g., in the case of concentration gradients in an osmotic cell or thermal gradients in a solid. In these cases, a steady-state condition only applies to a finite system or to an infinite system in the case of an infinite relaxation time. In this paper we investigate such phenomena by proposing a model[12] of a diffusive driven system which supports mass transport. The model, which is a kinetic lattice-gas model with repulsive interactions, is driven by external chemical potentials applied at the edges of a finite system. The model has been inspired by recent work on modeling of diffusion-controlled oxygen ordering in finite samples of high-T_c superconductors of the $YBa_2Cu_3O_{7-\delta}$ type.[13]

The kinetic lattice model is defined by (i) a Hamiltonian and (ii) a dynamical principle which embodies the imposed conservation laws. (i) The Hamiltonian is that of a nearest-neighbor repulsive lattice gas defined on a square lattice which is finite in one dimension and periodically infinite in the other direction. Chemical potentials μ_A and μ_B are applied at the rows of the two open ends A and B. The Hamiltonian reads

$$H = J\sum_{\langle i,j\rangle} n_i n_j - \mu_A \sum_{i \in A} n_i - \mu_B \sum_{i \in B} n_i, \qquad (1)$$

where $n_i = 0,1$ is the occupation variable of the ith site and $J > 0$. At low temperatures the pair interactions of the model support a simple (2×2) ordering. (ii) The dynamical principle, which strictly conserves the total density, chosen to be $\rho = \langle n_i \rangle = \frac{1}{2}$, makes the chemical potentials act like particle "pumps" at the system edges. Since the model has purely repulsive interactions, a conservation law for the density is necessary in order to stabilize ordered phases. Particles enter or leave a particular edge by particle-hole exchange with the opposite edge. The transition probabilities of this exchange only involve the pair interaction potential and not the chemical potentials. Consequently, the chemical potentials only enter the transition probabilities between the edge rows and the interior of the system. The dynamics of the interior of the lattice is that of Kawasaki nearest-neighbor and next-nearest-neighbor particle hopping. The total density (but not the order parameter) is hence conserved. It is important to note that by imposing these dynamics on the Hamiltonian via the particular particle

pumps, the system becomes conceptually different from that of an equilibrium system described by symmetry-breaking boundary fields which only influence the behavior over the range of the correlation length.[14] For $\mu_A \neq \mu_B$, the dynamics of the present model drives the system into a steady-state mass-transporting state which is a nonequilibrium state characterized by a net current of particles through the lattice and a nonuniform mass-density profile. The system is thermodynamically open since it requires an input of energy to lift particles up in the chemical-potential gradient across the system from one open edge to the other; e.g., the energy input is $\mu_B - \mu_A$ when a particle is moved from A to B. The steady state of the system is one in which this input of energy is balanced by the energy dissipated to the heat bath. As a result of the exclusion principle, the current vanishes in the thermodynamic limit, for all values of the chemical potential, as $Q \sim L^{-1}$, where L is the length along the direction of the current. It should be noticed, however, that for any finite value of L, the system will be driven into a steady-state transporting state after a sufficiently long equilibration time t. Hence, the steady state of the system survives provided the thermodynamic limit is taken in the order $t \rightarrow \infty$ and $L \rightarrow \infty$. Furthermore, the present setup in a finite geometry provides a simple means of directly measuring the transport properties of the system by choosing chemical-potential values corresponding to the linear-response regime.

The steady-state properties of the kinetic lattice model are studied by Monte Carlo computer-simulation techniques. The calculations are performed in a rectangular geometry for different lattice sizes. The case of asymmetric chemical potentials is considered, $\mu_A = -\mu_B = \mu$. The simulations are carried out for a series of different lattice sizes $L \times M$, where M denotes the length perpendicular to the current. As an operational criterion for the steady state we have specified that the flux measured at several parallel planes through the sample, including the edges, are the same within 0.5%. The statistics generated correspond typically to 10^5–10^6 Monte Carlo steps per site. The ensemble-average quantities calculated involve the mass-density profile $\rho(x)$, which is also averaged over the sites in the columns of the lattice perpendicular to the current, the energy fluctuations $\Delta E = \langle \mathcal{H}^2 \rangle - \langle \mathcal{H} \rangle^2$, the mass flux Q, and the column order parameter $\rho_s(x)$ (the ordering density). Furthermore, we have calculated the mass-transport coefficient[12] $D = Q(d\rho/dx)^{-1}$ in the cases where the mass-density profile is linear and one has Ohmic conductance. The various quantities are calculated as functions of chemical potential μ and temperature T.

In Fig. 1 are shown typical results for the mass-density profile $\rho(x)$ and the order-parameter profile $\rho_s(x)$ for a temperature $k_B T/J = 0.1$, which is low compared to the equilibrium transition temperature $k_B T_c/J \approx 0.56$. The data represent a 100×30 lattice. It is seen that for low values of μ the system is strongly ordered with an almost uniform $\rho_s(x)$ and with a mass-density profile which is almost constant but with a weak slope corresponding to a slight current. When μ increases there is a crossover to a quite different steady-state organization of the system in which the current effectively has destroyed the sublattice ordering over a substantial range of the system. As μ increases, this range approaches a temperature-dependent limit which is effectively reached in Fig. 1(c). This limit is characterized by a saturation of $\rho(x)$ at the two edges of the sample. These phenomena may be understood as follows: At zero temperature, due to the nearest-neighbor repulsion, there is a finite current in the system only for $\mu/J \geq 3$. Since the chemical potential is not conjugate to the order parameter, there is a progressive destruction of the sublattice ordering away from the edges. This leads to a nonlinear variation of $\rho(x)$ which may be seen as consisting of three different sigmoid segments which are merged at the two values of x which effectively correspond to the two percolation limits of $\rho(x)$, i.e., enclosing the range of densities, at a given temperature, which can support sublattice ordering.
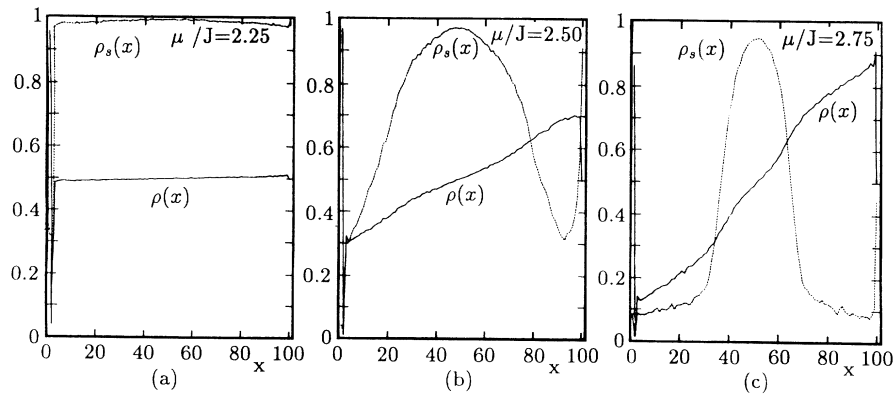

FIG. 1. Mass-density profile $\rho(x)$ and order-parameter profile $\rho_s(x)$ for a system with $L \times M = 100 \times 30$ sites, where L is the length in the transporting direction. Results are shown for three different chemical-potential values μ at a temperature $k_B T/J = 0.1$.

Hence, the steady-state system is driven into a heterogeneous phase-separating state in which a region with sublattice ordering is sandwiched between a gaseous low-ρ phase and a solid high-ρ phase. The phase-separated state is dynamically maintained. The shape of both profiles, $\rho(x)$ and $\rho_s(x)$, is only slightly sensitive to the size of the system and effectively scales with xL^{-1}. However, $\rho_s(x)$ has a measurable dependence on L in the center and tends to the equilibrium value as

$$\rho_s(x=L/2) \sim \rho_s^{L \to \infty}(x=L/2)(1-aL^{-1}) .$$

As the temperature is raised, the range of the sublattice ordering is diminished and above a certain temperature a perfectly linear mass-density profile establishes itself and only a finite-size rounded effect in $\rho_s(x)$ persists. The vanishing of the sublattice ordering proceeds via a nonequilibrium transition which will be analyzed below. Before we describe the properties of this transition, we show in Fig. 2 the characteristics of the current in the system as a function of temperature and chemical potential. Q is monotonously increasing with μ for all temperatures. This figure clearly demonstrates that, as the temperature is lowered, a clearer threshold is established below which only little current goes through the system. At $T=0$, Q is a step function jumping from $Q=0$ to $Q=7.20\times10^{-3}$ at $\mu/J=3$. Above this threshold, whose value depends on the details of the interaction potential, the current saturates as the chemical potential 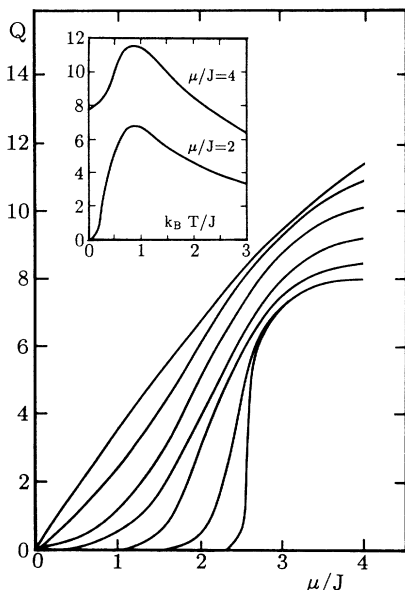is increased. The saturation is caused by the exclusion principle which only permits a finite transport capacity of a lattice system. The temperature variation of Q for values of μ above and below the zero-temperature threshold is shown in the inset of Fig. 2. It is seen that Q exhibits a maximum whose position is only slightly sensitive to the value of μ. The position of the maximum is at a temperature about twice as large as the equilibrium critical temperature.

Turning now to the phase behavior of the steady-state system, we are faced with the problem of identifying a suitable order parameter which characterizes the coherent order of the inhomogeneous system. For this purpose we have chosen the sublattice ordering at the center row of the system, $\Phi = \rho_s(x=L/2)$. In Fig. 3 is given a collection of data for $\mu/J=4$ in the case of a $100 \times M$ system, where M is varied in order to illustrate finite-size effects. When comparing with the corresponding order-parameter data for the equilibrium system, also given in Fig. 3, it is seen that the steady-state system is much more strongly influenced by finite-size effects. Similarly, the driven system is found to be subject to significantly stronger fluctuations in the transition region than the equilibrium system. Furthermore, these fluctuations increase as M is increased. At the same time the current is very stable. These observations indicate that the ordering process couples strongly to the current. This also implies that the sublattice order is significantly enhanced by the driving in the finite system as manifested in the high-temperature tails in Fig. 3. For given values of μ and L, the order-parameter curve approaches a limiting curve, as $M \to \infty$, which is significantly suppressed

FIG. 2. Current Q as a function of chemical potential μ. In increasing order, the different curves correspond to temperatures $k_BT/J=0.05$, 0.2, 0.3, 0.4, 0.5, 0.6, and 1.0. Inset: Current as a function of temperature for two different chemical-potential values. Q is in units of 10^{-3} Monte Carlo steps per site per lattice parameter.
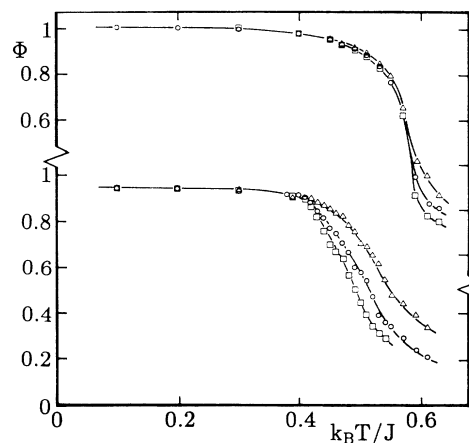

FIG. 3. Order parameter $\Phi(T)$ measured at the center row of the driven steady-state system for $\mu/J=4$. Results are shown for different system sizes $100 \times M$, where M is the length perpendicular to the transporting direction. $M=30$ ($\triangle$), 60 ($\bigcirc$), and 100 ($\square$). For comparison, results are also shown for an equilibrium system, i.e., $\mu=0$. The peculiar size dependence for the equilibrium system at temperatures below the transition is caused by the particular mixing of open and periodic boundary conditions.

below the equilibrium order-parameter curve. As $L \to \infty$ this limiting curve approaches the equilibrium, infinite-system order parameter. Finally, as μ is decreased, the limiting curve approaches that of the corresponding equilibrium situation.

The mass-transport coefficient $D(T)$ is found to be an increasing function of temperature in the Ohmic regime. Since the chemical potential is not conjugate to the order parameter, that is, $D(T)$ is not directly related to the ordering compressibility, $D(T)$ attains a nonzero value at the transition; i.e., the mass transport is not fully quenched by critical slowing down.[12,15] If we define the transition temperature $T_c(L,\mu)$ of the driven system as the temperature where the M dependence of Φ becomes negligible, we observe that for all finite values of L and μ the transition temperature of the driven system is suppressed below that of the corresponding equilibrium system. The present data are too imprecise to provide information about critical exponents.

Whereas the details of the results presented in the present paper are sensitive to the particular model studied, the general phenomena of steady-state mass transport in a finite nonequilibrium system driven by an external chemical-potential gradient are not. The phase transitions and the phase-separation phenomena in the driven system reflect the underlying phase behavior of the equilibrium system. Specifically, more complex phase-separation phenomena involving several phases can be triggered if further-neighbor attractive interactions are allowed for.[12] In the case of purely attractive nearest-neighbor interactions, a simple phase separation between a low-density phase and a high-density phase is found at low temperatures. In this case the current couples directly to the order parameter which is just the density. This situation has recently been studied by a time-dependent Ginzburg-Landau analysis[16] and it was suggested that the driven system is equivalent to an equilibrium system in a globally applied linear chemical potential proportional to the current. Computer simulations on the model qualitatively confirm this picture. If a similar analysis applies to the present model with repulsive interactions, it may be anticipated that a global linear chemical potential will provide a particular map of the equilibrium phase diagram. If such a picture turns out to be correct, it implies that it should be possible to probe phase equilibria by a kind of "imaging" technique involving transport in a finite-system geometry.

Finally, we wish to relate our findings to the properties of the driven diffusive system proposed by Katz, Lebowitz, and Spohn[1,2] and subsequently analyzed by a number of workers.[3-11] Firstly, our results apply to a system which is driven by only boundary fields, in contrast to the model by Katz, Lebowitz, and Spohn[1,2] which is driven by a long-range global field. Therefore, the effects of the drive on the phase transition in our model do not survive the thermodynamic limit, although the

long-range structural organization persists at low temperatures. Furthermore, the phase separation in our model is induced by the drive, whereas in the model by Katz, Lebowitz, and Spohn it is induced by simple symmetry breaking due to the conservation law imposed on the global density. An even more significant difference is that in our model there is a persistent inhomogeneity along the transporting direction. This latter fact makes it much more difficult to characterize and describe the cooperative phenomena of the model.

The present paper presents a first attempt to model nonequilibrium steady-state diffusive systems driven by local fields. Further work is required to obtain a more complete understanding of the phenomena. Such work could involve time-dependent Ginzburg-Landau analysis[16] or renormalization-group theory.[4,7]

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