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FAST-IONIC-CONDUCTOR BEHAVIOR OF DRIVEN LATTICE-GAS MODELS*

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We discuss driven diffusive lattice-gas systems as a model for fast ionic conductors, derive associated hydrodynamic equations and expressions for transport coefficients, and compare mean-field theory, Monte Carlo results and experimental observations. The comparison between model and real behaviours helps to understand some properties of those materials which seem to be characterized by the occurrence of nonequilibrium steady states and phase transitions. In particular, our study suggests the existence in Nature of a novel (nonequilibrium) universality class.

KEY WORDS: Superionic conductors, nonequilibrium phase transitions, driven diffusive systems, nonequilibrium steady states, hydrodynamic equations, nonequilibrium lattice models, statistical mechanics.

1 INTRODUCTION

Fast (or super) ionic conductors (FIC) are solid electrolytes. That is, whereas they may be insulators to the flow of electrons, FICs allow one type of ions to move in

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a liquid-like fashion through a solid matrix set up by other types of ions. Thus, in contrast to more familiar electrolytes, FIC do not need to be liquids, e.g. molten salts or aqueous solutions, to present a high ionic conductivity. Examples include inorganic crystals such as β- or β''-alumina, glasses such as $AgI\text{-}Ag_2MoO_4$, and polymers such as polyethylene oxide-$NaBF_4$ [see, for instance, Bates and Farrington 1981]. They have been very actively studied for more than a decade (see, for instance, Sato and Kikuchi, 1971; Boyce and Huberman, 1979; Salomon, 1979; Dieterich *et al.*, 1980; Bates *et al.*, 1982; Olson and Adelman, 1985; den Hartog and van der Veen, 1988, and references therein). In fact, such a diversity of materials is most suitable for many technological applications (e.g. high-temperature batteries, fuel cells, ion-selective membranes, gas sensors and other electrochemical devices), and a full understanding of their properties involves some challenging fundamental problems in solid state and statistical physics (e.g. those related to diffusion, atomic mechanisms of migration, and nonequilibrium steady states and phase transitions).

Experiments on FIC materials have revealed, in particular, that the ionic-conductivity versus temperature curve usually allows one to characterize two well-defined phases below the melting point, namely a low-temperature poor ionic-conductor phase and a good ionic-conductor phase at a relatively higher temperature. This and some other observations reveal that FIC may undergo a nonequilibrium phase transition influenced, perhaps even dominated, by the external electric field which produces the (dissipative) steady current of ions (Katz *et al.*, 1983; 1984; Marro *et al.*, 1984; Vallés and Marro, 1986). Fortunately, the understanding of these and other facts concerning FIC behaviour seems to be one of those rare practical problems in physics where a transition from microscopic to macroscopic levels of description is feasible at present. In order to illustrate this, we study here the properties of FIC materials by looking at the behavior of some related microscopic models (see, for instance, Katz *et al.*, 1983; Marro *et al.*, 1984; van Beijeren and Schulman, 1984; Vallés and Marro, 1986, 1987; Krug *et al.*, 1986; Leung and Cardy, 1986; Marro *et al.*, 1987; Wang and Lebowitz 1988; Garrido and Marro, 1989; Garrido *et al.*, 1990a). Namely a series of models originally developed to deal with more general problems in statistical mechanics. In particular, we shall characterize the steady states and phase transitions occurring in the models, refer to some existing experimental data, and try to encourage experiments. These may be decisive in producing a better classification of materials, and in confirming the existence of a novel, nonequilibrium universality class thus evidencing the basic ingredients determining FIC macroscopic behavior.

The paper is organized as follows. Section 2 contains a critical description of driven diffusive lattice-gas models. Section 3 describes mean-field solutions and Monte Carlo computations globally producing an interesting characterization of steady states, phase transitions and critical phenomena. The macroscopic description is fulfilled in Section 4 with a derivation of related hydrodynamic-like equations and transport coefficients. The picture emerging from Sections 3 and 4 is shown to be qualitatively consistent with some representative observations on real materials. Our main conclusions are summarized in Section 5.

2 THE BASIC MODEL SYSTEM

Diffusion problems may sometimes be clarified by simple random walk studies. Nevertheless, when the jump of the ions is greatly influenced by their immediate surroundings, as may occur in the diffusion in FIC systems (Sato and Kikuchi, 1971), the study of lattice-gas models is more convenient. After some pioneering studies, where often the microscopic details were unnecessarily stressed and/or the apparent nonequilibrium nature of the phenomena was underestimated, the so-called driven diffusive lattice-gas models have been studied systematically in an effort to gain a better understanding of several fundamental problems. Namely, to investigate steady states and their associated instabilities (Katz *et al.*, 1983, 1984; Marro *et al.*, 1987; Wang and Lebowitz, 1988; Garrido and Marro, 1989), to obtain macroscopic equations from microscopic models (see, for instance, the review by Lebowitz *et al.*, 1988; see also Garrido *et al.*, 1989) and, in the long run, to construct a unified nonequilibrium statistical mechanics. Those models have been studied by means of Monte Carlo simulations (Marro *et al.*, 1984; Vallés and Marro, 1986, 1987; Marro and Vallés, 1987; Marro *et al.*, 1987), mean-field theory (van Beijeren and Schulman 1984; Krug *et al.*, 1986; Dickman, 1988; Garrido *et al.*, 1990a), and field-theoretical methods (Gawedzki and Kupiainen, 1986; Janssen and Schmittmann, 1986; Leung and Cardy, 1986); there are also some studies of interfaces (Leung, 1988; Hernández-Machado and Jasnow, 1988; Leung *et al.*, 1988, 1989; Vallés *et al.*, 1989; Hernández-Machado *et al.*, 1989) and correlations (Vallés and Marro, 1987; Zhang *et al.*, 1988; Garrido *et al.*, 1990b). In this paper we shall consider the basic model in those studies because it also serves to illustrate microscopic and macroscopic features of FIC materials.

Consider a simple cubic lattice in d dimensions having $N = L^d$ sites, and assume periodic boundary conditions. The occupation variable at each site $\mathbf{x}$ has two different states, $\sigma_{\mathbf{x}} = 1, 0$, which represent respectively the presence of a particle (to be interpreted as a positive ion) at $\mathbf{x}$, and $\mathbf{x}$ being an empty site. That is, the system has 2^N different configurations, each containing $nN \equiv \sum_{\mathbf{x}} \sigma_{\mathbf{x}}$ particles and $(1-n)N$ holes. In addition to the repulsive core implied by that exclusion principle, the particles interact according to the Hamiltonian

$$H(\sigma) = -4J \sum_{|\mathbf{x}-\mathbf{y}|=1} \sigma_{\mathbf{x}}\sigma_{\mathbf{y}}, \tag{2.1}$$

for instance. Here, $\sigma \equiv \{\sigma_{\mathbf{x}}\}$ and the sum is over nearest neighbor (*nn*) pairs. There is also an external uniform electric field, E, along one of the principal directions of the lattice which, together with the (stochastic) action of a heat reservoir at temperature T, induces the system time evolution according to the Master Equation:

$$dP_E(\sigma; t)/dt = \sum_{|\mathbf{x}-\mathbf{y}|=1} [c_E(\sigma^{\mathbf{x},\mathbf{y}}; \mathbf{x}, \mathbf{y})P_E(\sigma^{\mathbf{x},\mathbf{y}}; t) - c_E(\sigma; \mathbf{x}, \mathbf{y})P_E(\sigma; t)]. \tag{2.2}$$

Here, $\boldsymbol{\sigma}^{\mathbf{x},\mathbf{y}}$ represents the configuration σ with the occupation variables at *nn* sites $\mathbf{x}$ and $\mathbf{y}$ exchanged, $P_E(\sigma; t)$ is the probability density of configuration σ at time t, and

$c_E(\sigma; \mathbf{x}, \mathbf{y})$ is the transition probability per unit time of having that exchange when the system configuration is σ.

In the absence of the field this is the familiar kinetic lattice-gas or Ising model with Kawasaki (1972) dynamics. As it is well-known, this leads, for any transition probability satisfying detailed balance, to an equilibrium state for each n specified by

$$P_{eq}(\sigma) = \exp[-\beta H(\sigma)]\{\textstyle\sum_\sigma \exp[-\beta H(\sigma)]\}^{-1}, \tag{2.3}$$

where $\beta = 1/k_B T$. For $n = \frac{1}{2}$, this system is known to undergo (in the infinite volume limit) a second-order phase transition at critical temperature $T_c^0 k_B/J = 0$, 2.27 and 4.5 for $d = 1$, 2 and 3 respectively, i.e. there is a unique state for $T \geq T_c^0$ while there is (isotropic) phase segregation for $T < T_c^0$, $d > 1$. The field **E**, however, induces a preferential hopping in the field direction. This leads, for periodic boundary conditions in that direction, to a nonequilibrium steady state $P_E(\sigma)$ with a net current whose associated energy is absorbed by the involved heat reservoir. The bias is most naturally induced by adding to the energy difference $\delta H = H(\sigma^{x,y}) - H(\sigma)$ the work done by the field during a jump. This yields the transition probabilities per unit time for the exchanges between $\mathbf{x}$ and $\mathbf{y}$, $|\mathbf{x} - \mathbf{y}| = 1$, as

$$c_E(\sigma; x, y) = \Phi[\beta \delta H - \mathbf{E} \cdot (\mathbf{x} - \mathbf{y})(\sigma_x - \sigma_y)]. \tag{2.4}$$

Here, Φ represents any function such that $c_E(\sigma; \mathbf{x}, \mathbf{y})$ satisfies the usual condition of detailed balance when $E = 0$. For instance,

$$\Phi(h) = \min[1, e^{-h}] \tag{2.5a}$$

or else

$$\Phi(h) = [1 + e^h]^{-1} \tag{2.5b}$$

which, for $E = 0$, correspond respectively to the rates first introduced by Metropolis *et al.* (1953) and by Kawasaki (1972) in the study of equilibrium problems. By contrast, (2.4) for $E > 0$, which makes $P_E(\sigma)$ invariant under translations and symmetric under the transformation $E \to -E$ and $\sigma_x \to 1 - \sigma_x$ for all $\mathbf{x}$, only satisfies a detailed balance condition locally (Katz *et al.*, 1984).

The nonequilibrium stationary state $P_E(\sigma)$ generated by the above dynamics has no trivial relation with $P_{eq}(\sigma)$. On the one hand, $P_E(\sigma)$ cannot in general be described by a formula of type (2.3) for any "reasonable", e.g. short-ranged, Hamiltonian [Garrido and Marro 1989]. That is, because of periodic boundary conditions, the uniform field E is not the gradient of a potential, so that the electric energy in particular cannot be included in the Hamiltonian and there is no stationary solution of (2.2) such as (2.3). On the other hand, contrary to the equilibrium case as long as detailed balance is satisfied, the nonequilibrium steady-state distribution will depend in general (Singer and Peschel, 1980; Katz *et al.*, 1984; Vallés and Marro, 1986; Garrido and Marro, 1989) on the specific rate (2.4) since that is only defined as the stationary solution of a certain Master Equation. It may be mentioned, however, that there is some evidence, both from analytical (Krug *et al.*, 1986; Lebowitz *et al.*, 1988; Dickman, 1990) and Monte Carlo (Marro *et al.*, 1984; Vallés and Marro, 1986; Zhang, 1987; Wang *et al.*, 1989) studies (see also below), that most

features of the phase diagram and the critical exponents are rather insensitive to the specific rates under some (but not all) circumstances; in particular, that seems the case in the present problem. Those two facts evidence that the study of the (nonequilibrium) steady states implied by Eqs (2.2) and (2.4) requires a non-trivial generalization of the usual methods in equilibrium statistical mechanics. The latter, however, may still serve as a reference in some cases. For instance, one may show that Eqs (2.2) and (2.4) imply the Kubo formulae for bulk diffusivity and electrical conductivity (Katz *et al.*, 1984). It may also be shown that the state of minimum entropy production (de Groot and Mazur, 1984; Nicolis and Prigogine, 1977; Haken, 1983) only agrees with $P_E(\sigma)$ to linear order in E (Katz *et al.*, 1984).

The model representation of the actual interactions between ions also deserves a comment. Due to a familiar screening (collective) effect, the interactions may be assumed to be reduced in practice to a hard-core repulsion plus an effective *short-ranged* interaction which is negelected beyond *nn* positions. It may be argued [see, for instance, Sato and Kikuchi, 1971; Murch and Thorn, 1977] that the latter should be repulsive or "antiferromagnetic", i.e. $J < 0$ in Eq. (2.1), in order to agree better with the behavior of FIC materials. Nevertheless, we shall refer in the following to both, $J < 0$ and $J > 0$, and our analysis seems to indicate that the latter case (i.e. attractive or "ferromagnetic" *short-ranged* interactions) is the most relevant one for understanding FIC behavior.

Previous studies (Katz *et al.*, 1984, Garrido *et al.*, 1990a) indicated that, as the field strength increases, both the interesting new features of the stationary state get more pronounced and the magnitude of the net current saturates for some value of the field. Therefore, that of a strong field is the most relevant and also the most suitable case for theoretical analysis; namely a saturating field, or theoretically $E \to \infty$, which simply means that motion contrary to the field is forbidden. It is true that some experiments refer to relatively small electric fields; nevertheless, they may be well within the saturation regime, and it seems that varying E in the model with $J > 0$ only produces some quantitative differences.

It should also be remarked that we shall mainly refer to the cases $d = 1$, 2. This is dictated by the fact that the situation for $d = 3$ is much less clear cut at present, and also because low dimensional effects are very relevant in FIC materials. That is, a number of real compounds have layer or channel structures which suggest that the conductivity is confined to one or two dimensions (Richards, 1977, 1978). For instance, the sodium ions in β''-alumina (Kennedy, 1977) and the silver ions in $AgCrS_2$ (Brüesch *et al.*, 1983; Beyeler, 1981) basically remain within lattice planes, β-eucryptite ($LiAlSiO_4$) (Alpen *et al.*, 1977) and potassium hollandite ($K_{1.54}Mg_{0.77}Ti_{7.23}O_{16}$) (Bernasconi *et al.*, 1979) are quasi one-dimensional conductors where ions are compelled to move in channels, and organic conductors and/or semiconductors like tetrathiafulvalen-tetra-cyanoquinodimethane (TTF-TCNQ) display highly anisotropic conductivity (see, for instance, Heeger *et al.*, 1975). Thus, $d = 1$ and $d = 2$ models have more than an academic interest.

Finally, it may be emphasized as a general remark that most observable behavior such as critical phenomena and hydrodynamic laws is determined by some very general microscopic features and by the existence of very different spatial and

temporal scales at the microscopic and macroscopic levels of description. Consequently, microscopic models can be rather crude, even apparently wrong, and still give rise to correct macroscopic behavior. All that is necessary is that they contain the basic qualities responsible for the phenomena of interest, a condition which seems to hold in the present case. As a matter of fact, the claim that an oversimplified model may capture many essential physical features of FIC is indorsed by the observation that quite different materials, including inorganic crystals, glasses, polymers, etc., seem to present the same basic phenomena.

3 MACROSCOPIC BEHAVIOR

This section describes some results obtained by two main approaches, namely, mean-field techniques for $d = 1$ and Monte Carlo (MC) computations for $d = 2$ and 3. The ensuing macroscopic description is compared with the one in other approaches on similar models and with some typical observations in FIC materials.

3.1 *Mean-field approach for* $d = 1$

The model has been studied analytically under some limiting conditions. Namely, when $E \to \infty$ and the ratio, Γ, of exchanges attempted along the direction of the field to those transverse to the field is infinite (instead of $\Gamma = 1$ as in the basic version) (van Beijeren and Schulman, 1984; Krug *et al.*, 1986). That situation is of great theoretical interest since it produces a model that is exactly solvable above the critical temperature with non-trivial behavior. That is, the limit $\Gamma \to \infty$ compels the particles to move along chains, as in an assembly of one-dimensional Ising models with interactions between neighboring chains which influence the dynamics and lead to the possibility of a phase transition. The practical relevance of such a significant modification of the model is more questionable, however. Consequently, we shall focus instead on a cluster-variation method of solution for $\Gamma = 1$ which is also applicable to other dynamical problems.

The microscopic functions $A(\sigma)$ leading to relevant macroscopic quantities $\langle A \rangle_t$ usually depend on the occupation variables at a relatively small, compact set of lattice sites, say a local domain D. That is, $A(\sigma) = A(\sigma_D)$ where σ_D represents the configuration within the domain D, and

$$\langle A \rangle_t = \sum_{\sigma} P(\sigma; t) A(\sigma) = \sum_{\sigma_D} Q(\sigma_D; t) A(\sigma_D) \tag{3.1}$$

where $Q(\sigma_D; t) \equiv \sum_{\sigma \sim \sigma_D} P(\sigma; T)$. Thus, one may combine (3.1) with (2.2) to obtain specific equations for the time evolution of mean local quantities by neglecting net currents through the surface of D and, consequently, some local fluctuations. The nature of the resulting approximation depends on the choices for both, D and $Q(\sigma_D; t)$ (Garrido *et al.*, 1990a). For instance, one may write $Q(\sigma_D; t)$ in (3.1) as a function of (only) the system densities of *nn* pairs $+ +$, $+ -$ and $- -$ (where $-$ and $+$ represent respectively a hole and an occupied site) to be denoted respectively by u, e, and z; $u + e + z = 1$. Thus, when D is of minimum size (say its interior just contains enough sites to allow for the basic dynamical process) our approximation essentially corresponds to that of Bethe–Peierls, i.e. a first-order mean field description [see, for instance, Ziman 1979]. Notice that, as usually occurs for nonequilibrium phenomena, mean-field approaches are expected to be more realistic here than in the study of equilibrium phenomena. As a matter of fact, the model with E and Γ infinite was shown (van Beijeren and Schulman, 1984; Krug *et al.*, 1986) to exhibit a critical point of the mean-field Ising variety, and MC computations (Vallés and Marro, 1987) described a crossover towards mean-field behavior with increasing Γ.

Consider the case $d = 1$, i.e. the lattice forms a large closed chain under the action of a uniform field, $E = E\hat{\mathbf{x}}$, where $E > 0$ and $\hat{\mathbf{x}}$ defines a positive direction along the chain. The dynamics consists of *nn* exchanges, so that the particle density $n = u + e$ is conserved, which are performed according to a transition probability per unit time given by $w(\delta H) = \frac{1}{2}[\Phi_+ + \Phi_-]$ where $\Phi_\pm = \{1 + \exp[\beta\delta H \pm E]\}^{-1}$, i.e. the choice (2.5b), for instance. This amounts to considering separately, with equal a priori probabilities, $\Phi_\pm$ respectively, the possibility of jumps in the directions $\pm\hat{\mathbf{x}}$ biased with a probability which favors the jumps along the field direction and depends on the change in the system energy produced by the attempted jump.

Under the present conditions, the minimum allowable domain D suffices to produce a satisfactory description. It consists of four consecutive lattice sites where the interior pair is the only one available for exchanges. Accordingly, the relevant macroscopic variable is the density of *nn* pairs of particles, u, whose time evolution for the choice (2.5b) simply follows (Garrido *et al.*, 1990a) by the method outlined above as $du/dt = F(u; n, T, E, J)$ with

$$F = \frac{2(n-u)}{n(1-n)}\left[(n-u)^2 w(-4J) - u(1-2n+u)w(4J)\right] \tag{3.2}$$

and $w(\pm 4|J|) = \frac{1}{2}\{[1 + \exp(\pm 4\beta|J| - E)]^{-1} + [1 + \exp(\pm 4\beta|J| + E)]^{-1}\}$; this is valid for both, $J > 0$ (attractive ion interactions) and $J < 0$ (repulsive interactions). The general solution of that equation for u is

$$\frac{[2\delta(n-u) + w(4J) - \chi]^{\alpha^+}}{[2\delta(n-u) + w(4J) + \chi]^{\alpha^-}}\,[2n(1-n)(n-u)^2\delta]^{-1} = \exp(-4t + C_0) \tag{3.3}$$

where $\delta \equiv w(-4J) - w(4J)$, $\chi \equiv [w(4J)^2 + 4n(1-n)\delta w(4J)]^{1/2}$, $\alpha^+ \equiv w(4J)/\chi \pm 1$, and C_0 is a constant related to the initial conditions. One also has, for instance, the special solutions

$$u = n - (n - u_0)(at + 1)^{-1/2}, \quad u_0 \equiv u(t = 0), \tag{3.4}$$

where $a = 4(n-u_0)^2[n(1-n)]^{-1}w(-4J)$, when $w(4J) = 0$ and $w(4J) > 0$,

$$u \approx [4w(4J)t]^{-1} \tag{3.5}$$

when $w(4J) > 0$, $w(-4J) = 0$ and $n = \frac{1}{2}$, and

$$u = n - n(1-n)\{1 - [(n^2 - u_0)/(n - u_0)]\exp[-2w(4J)t]\}^{-1} \tag{3.6}$$

when $w(4J) = w(-4J) > 0$. Thus, the system may show up an exponential decay,

(3.6), or a slower relaxation as in (3.4) and (3.5), the latter corresponding to the presence of a critical point at $T = 0$.

The steady states follow from above as $t \to \infty$. We get respectively from (3.3)–(3.6) that

$$u_{st} = n + \tfrac{1}{2}[w(4J) \pm \alpha]\delta^{-1}, \tag{3.7}$$

$$u_{st} = n, \quad \text{for} \quad T \to 0,\ J > 0,\ \text{finite } E, \tag{3.8}$$

$$u_{st} = 0,\ 2n - 1, \quad \text{for } T \to 0,\ J < 0,\ \text{finite } E,\ n \geq \tfrac{1}{2}, \tag{3.9}$$

$$u_{st} = n^2, \quad \text{for all} \quad J, \tag{3.10}$$

either as $T \to \infty$ for all finite E or as $E \to \infty$ for all T.

A necessary condition for the stability of the corresponding stationary solution, $F(u_{st}) = 0$, is that

$$(dF/du)_{u=u_{st}} < 0, \tag{3.11}$$

and the critical condition $(dF/du)_{st} = 0$ may indicate the onset of a (nonequilibrium) phase transition. Thus, stability requires that

$$3\delta(n - u)^2 + w(4J)[n(1 + n) - 2u] > 0. \tag{3.12}$$

It follows that only the negative sign in (3.7) produces a stable solution, that (3.10) is always stable, that (3.8) has critical stability, i.e. the expression in (3.12) equals zero when $u = n$ and $w(4J) = 0$, and that the two solutions (3.9) are only stable for certain values of n, i.e. $u_{st} = 0$ is stable for $n < \frac{1}{2}$ and has critical stability when $n = \frac{1}{2}$, while $u_{st} = 2n - 1$ is stable for $n > \frac{1}{2}$.

Most interesting is the net current induced by the external electric field $E\hat{\mathbf{x}}$. That may be defined as

$$j(t;\ T,\ E,\ n) = \lim_{\delta t \to 0} (N, \delta t)^{-1}[N_{\mathbf{x}}(t,\ \delta t) - N_{-\mathbf{x}}(t,\ \delta t)] \tag{3.13}$$

where N is the number of particles in the system, and $N_{\pm\mathbf{x}}(t, \delta t)$ represents the number of particles hopping to their nn empty positions in the direction $\pm\hat{\mathbf{x}}$ between times t and $t + \delta t$. The explicit computation of (3.13) for the dynamics (2.5b) produces:

$$j(t;\ T,\ E,\ N) = \frac{2(n-u)}{n(1-n)} \sinh(E)\left\{\frac{(n-u)(1-2n+2u)}{2(1+\cosh(E))}\right.$$
$$\left. + [u(1-2n+u) + (n-u)^2][(e^{-4\beta|J|} + e^{-E})(1 + e^{4\beta|J|+E})]^{-1}\right\} \tag{3.14}$$

for all J and E; $u = u(t)$. The infinite field limit is $j(t;\ T,\ \infty,\ n) = n - u(t)$, i.e. it is simply related to the behavior of the energy. Several other limiting conditions may be worked out from those expressions as well; it follows in particular that limits concerning T and E, for instance, do not commute in general. On the other hand, the stationary values of the current, j_{st}, easily follow by combining (3.14) with Eqs. (3.7)–(3.10) for u_{st}.

The resulting behavior is illustrated by Figures 1 and 2. Figure 1 represents $j_{st}(E)$ for $J > 0$; the case $J > 0$ is qualitatively similar. Figure 2 represents $j_{st}(T)$ for $J > 0$;

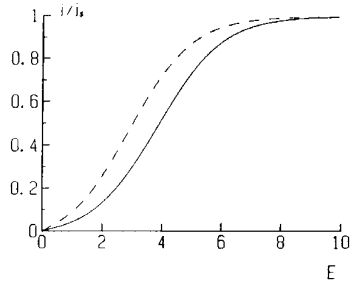


Figure 1 Variation with field strength E and system density n of the steady current, normalized to its saturation value, for $T = Jk_B^{-1}$ in the one-dimensional mean-field model for attractive short-ranged interactions ($J > 0$); the solid and dashed lines are for $n = \frac{1}{2}$ and $n = 0.1$ respectively.

the case $J > 0$ has, independently of n, a behavior qualtiatively similar to the one shown by the graph for $n = \frac{1}{2}$ in Figure 2. The situation depicted by those figures is most interesting. In particular, it suggests that the one-dimensional system presents two qualitatively different, well-defined behaviors. The first one, for $J < 0$ and $n \neq \frac{1}{2}$, is characterized (as illustrated for $E = 10$ by the two graphs near the top in Figure 2) by good conductor states both at low and high temperatures, with however a sudden current increase around $T = 4/E$. The second behavior, for $J < 0$ and $n = \frac{1}{2}$ (see the bottom graph in Figure 2) and for $J > 0$ at any density, is characterized by a continuous crossover from an insulator state when $T \ll 4/E$, to a good conductor state when $T > 4/E$. The first behavior is clearly a consequence of the degeneracy of the ground "antiferromagnetic" ($J < 0$) states when $n < \frac{1}{2}$, a fact which favors large stationary currents at low temperatures. Although we never found a similar behavior

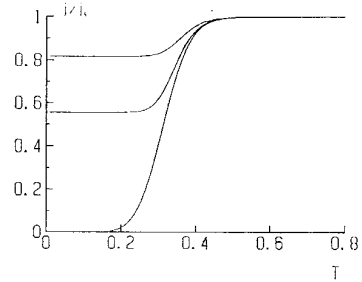


Figure 2 Same as Figure 1 showing the variation with temperature, in units of $|J|k_B^{-1}$ for repulsive interactions ($J < 0$) and, from top to bottom, $n = 0.3$, 0.4 and $\frac{1}{2}$. The latter graph may also represent the qualitative behavior when $J > 0$.

described in the relevant literature, we believe this should be an observable effect; in particular, real quasi-one-dimensional FIC should probably reveal qualitatively different behaviors depending on the sign of their effective ionic interactions.

The stationary current for small fields may be obtained from (3.14) in the limit $t \to \infty$ after performing an expansion around $E = 0$. It then follows the *electric conductivity* as $s \equiv (dj_{st}/dE)_{E=0} = e^{-2\beta|J|}[2(1 + e^{-4\beta|J|})(1 + e^{-2\beta|J|})]^{-1}$ for $J < 0$ and $n = \frac{1}{2}$, and the relation $s(J > 0) = \exp[-2\beta|J|]s(J < 0)$. That is, the conductivity is enhanced when the effective interactions between the ions are repulsive. Figure 3 represents the behavior of $s(K)$ with $K \equiv J/k_B T$ for the dynamics (2.5a) and (2.5b), and provides a comparison with some Monte Carlo data for the same model (Katz *et al.*, 1984) and with the analytical solution of a related model (Dieterich *et al.*, 1980); this reveals in particular a great consistency between the various approaches.

The system configurational energy per lattice site implied by the Hamiltonian (2.1) is $\langle e \rangle = \langle H/N \rangle = -4Ju$ where u may be interpreted either as the stationary value u_{st} or as a time dependent quantity $u(t)$, both given before. During the stationary regime, one may define a (nonequilibrium) specific heat as

$$C \equiv d\langle e \rangle_{st}/dT = -(4J/T^2)[du_{st}/d\beta] \tag{3.15}$$

whose behavior is illustrated by Figure 4 for $J > 0$ and dynamics (2.5b). A different choice of dynamics and/or interactions essentially produces the same qualitative behavior. Figure 4 reveals in particular that the maximum characterizing the equilibrium state ($E = 0$) increases and shifts towards smaller temperatures as E is increased, that the maximum location is at $\beta_m \approx E/4$ for large enough fields, i.e. around the temperature locating the transition from insulator to good conductor

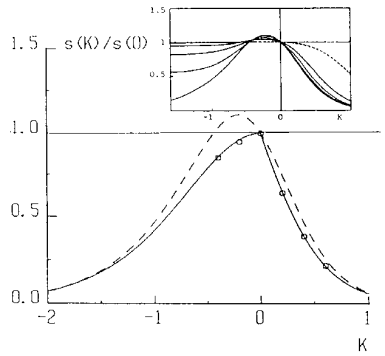


Figure 3 Variation with $K \equiv J/k_B T$ of the electric conductivity for the system in Figure 1 with rates (2.5a) (solid line) and (2.5b) (dashed line) when $n = \frac{1}{2}$. The circles are Monte Carlo data for the same model with dynamics (2.5a) (Katz *et al.*, 1984). The inset depicts analytical results with varying n in a related model (Dieterich *et al.*, 1980).

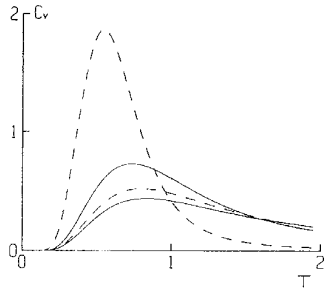


Figure 4 The specific heat (3.17) for the one-dimensional system with rates (2.5b) and attractive interactions, as a function of temperature and, from bottom to top, for increasing values of the field ($E = 0$, 1, 2 and 5 respectively).

that we have described above, and that C will essentially develop a Dirac delta function shape at $T = 0$ as $E \to \infty$.

Finally, one may also compute correlation functions such as the structure function $S(k)$, or Fourier transform of the pair correlation function $G(j) = \langle \sigma_0 \sigma_j \rangle - n^2$, with the philosophy of the present method. For the infinite ($N \to \infty$) system, that follows as

$$S(k) = 2n(1-n)\frac{\delta \cos(k) - \delta^2}{1 + \delta^2 - 2\delta \cos(k)}. \tag{3.16}$$

Notice that the long-range order limits, $S(k = 0)$ for $J > 0$ and $S(k = \pi)$ for $J < 0$, diverge as $\delta \to \pm 1$ respectively, i.e. there is a critical point at $T = 0$ in the system with attractive interactions, where $u = n$, and also for repulsive interactions when $n = \frac{1}{2}$; otherwise ($n = \frac{1}{2}$ and $J < 0$) the ground state degeneracy precludes long-range order at any temperature, excluding the trivial cases $n = 1, 0$.

3.2 *Monte Carlo results for $d = 2$*

The two-dimensional model has been the subject of more studies by Monte Carlo (MC) methods for both types of interactions, $J > 0$ (Vallés and Marro, 1986, 1987; Marro and Vallés, 1987) and $J < 0$ (Leung *et al.*, 1989). Most simulations for $d = 2$ refer to the rates (2.5a), to the basic choice $\Gamma = 1$, and to the case of a saturating field. We summarize here the results which we feel most relevant to guide observations on FIC materials, in particular those concerning the limit $N \equiv L^2 \to \infty$ as obtained from the study of several lattice sizes (L ranging from 10 to 300 in the actual MC experiments).

The study of attractive short-ranged interactions ($J > 0$) seems, even *a priori*, more relevant in relation with FIC behavior. That is, $J > 0$ already involves a strong repulsion, that forbidding two or more ions from occupying the same site, and it

may be that this is the most essential repulsive attribute of the interactions needed to obtain FIC behavior. The main qualitative conclusion for $J > 0$ concerns the existence for all field strengths of a nonequilibrium (anisotropic) phase segregation below a transition temperature, $T_c(E)$, which is an increasing function of E with an upper bound T_c^∞ for saturating fields. The nature of the phase transition changes as one varies the density, $n = N^{-1}\sum_x \sigma_x$. That is, the phase transition is continuous or second order, with a critical point at $T_c^\infty > T_c^0$ (the equilibrium, Onsager critical temperature), when the lattice is half-filled with ions ($n = n_c = \frac{1}{2}$), while it is discontinuous or first-order for $n \leq 0.2$, and the discontinuities, if any, are very weak within the range $n_c > n > 0.35$. The resulting phase diagram is represented in Figure 5.

The MC experiments also reveal that, while the phase segregation when $E = 0$ occurs in randomly oriented regions, the segregated states for $E \neq 0$ are highly anisotropic, e.g. they consist of a fluid or particle-rich phase where particles are clustered into a single strip (parallel to the field direction $\hat{\mathbf{x}}$) which coexists with a gas or particle-poor phase filling the rest of the system homogeneously. Figure 6 represents some typical configurations. The present situation may be understood by simply extrapolating some equilibrium theory to the case of a nonuniform system. That is, the system forms the minimum number of interfaces (one) which are consistent with the given (periodic) boundary conditions in order to minimize the total interfacial free energy. Also interesting is the fact that, while the system is essentially homogeneous above T_c^∞, where it shows up in a gas phase, a detailed study of the configurations uncovers, even well above T_c^∞, weak power-law decaying correlations corresponding to some anisotropic clustering mainly directed along the field direction; cf. (Vallés and Marro, 1987; Zhang *et al.*, 1988) for more details. One would expect this to produce some interesting experimentally observable facts for $d \geq 2$, namely, non-analyticities of the static structure function $S(\mathbf{k})$ near the origin $\mathbf{k} = 0$

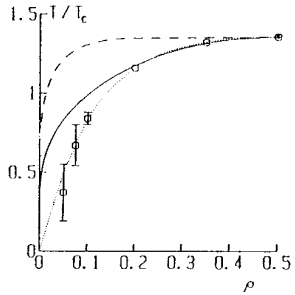


Figure 5 Variation with density of the transition temperature, and corresponding "error bars" locating the limit of metastability or spinodal line, as obtained by the Monte Carlo method for the system $d = 2$, $\Gamma = 1$, Metropolis dynamics (2.5b), attractive short-ranged interactions, and a saturating electric field. The solid line is the mean-field result and the upper dashed line is the equilibrium Onsager result, both normalized to the equilibrium critical temperature.

and self-similarity (Marro *et al.*, 1979; Marro and Toral, 1986) of the dynamical structure function $S(\mathbf{k}, t)$ even above the critical temperature.

A more quantitative picture of the phase segregation can be obtained from the behavior with n and T of an appropriate order parameter. This may be defined, for instance, as

$$m = \tfrac{1}{2}[n(1-n)]^{1/2}[\langle M_x^2\rangle_T - \langle M_y^2\rangle_T]^{1/2} \tag{3.17}$$

where $\langle\cdot\rangle_T$ denotes the ensemble average at temperature T produced by the

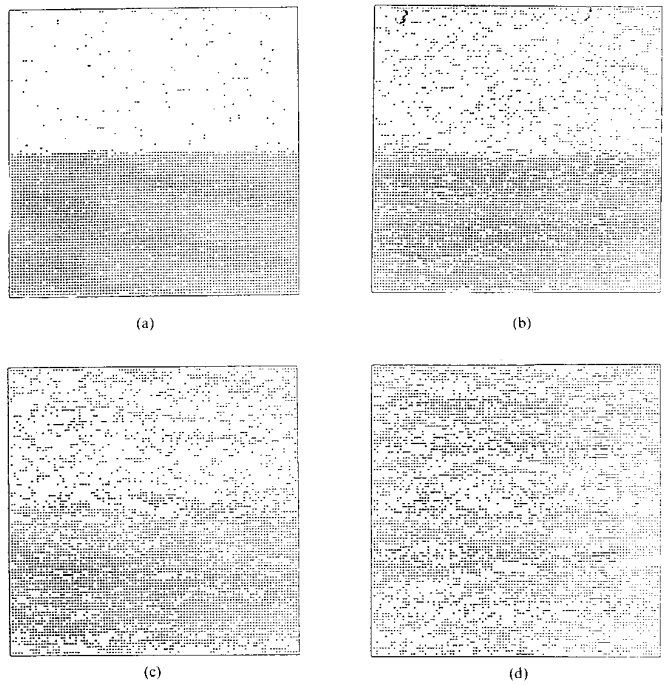
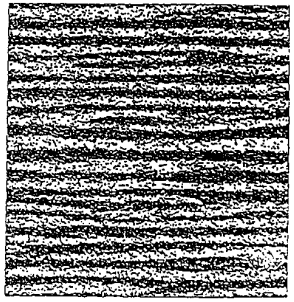
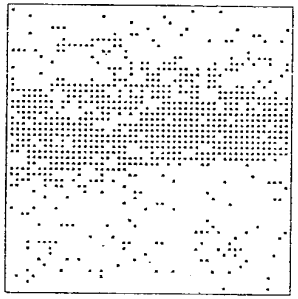


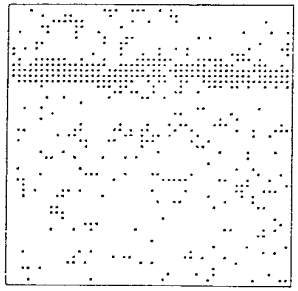
Figure 6 Some MC configurations of the two-dimensional system under a large horizontal electric field. **(a)** Typical steady state configuration for $n = \frac{1}{2}$ and $T = 0.8T_c^0$ ($L = 100$). (b) Same for $T = 1.2T_c^0$. **(c) Same for $T = 1.3T_c^0$, still below the critical temperature T_c^∞ induced by the field.** (d) Same for $T = 1.45T_c^0 > T_c^\infty$. (e) Typical intermediate multi-strip state for $T = 1.1T_c^0$ ($L = 300$); this never decayed into a one-strip state during the simulations. (f) Typical steady state configuration for $n = 0.35$ and $T = 1.2T_c^\infty(n)$ ($L = 50$). (g) Same as (f) for $n = 0.2$ and $T = 1.13T_c^0$.



(e)



(f)



(g)

Figure 6 *(continued)*

dynamics (2.4), and M_x^2 and M_y^2 represent respectively the squared longitudinal and transversal "magnetizations", say

$$M_{x(y)}^2 = L^{-1} \sum_{y(x)} \left[L^{-1} \sum_{x(y)} (2\sigma_{xy} - 1) \right]^2, \tag{3.18}$$

with $\sigma_{xy} \equiv \sigma_x$, where x and y describe the two principal lattice directions. The quantity m represents some measure of the differences between fluid (strip-like) and vapor (isotropic) phases; in particular, $\langle M_x^2 \rangle = \langle M_y^2 \rangle$ in the limit of infinite temperature as $N \to \infty$, implying $m = 0$, while $\langle M_x^2 \rangle \to 1$ and $\langle M_y^2 \rangle \to 0$ in the limit of zero temperature, and consequently $m \to 1$. Figure 7 depicts the behavior of $m(n, T)$; this confirms previous statements, e.g. m reveals itself as a continuous function of temperature for $n = \frac{1}{2}$ and perhaps also for $n = 0.35$, while it has a clear discontinuity for $n \leq 0.2$. Even though the study of m should suffice to reveal most important facts in the present problem with a particle conserving dynamics, one may attempt a more global description of order; for instance, by considering all possible Fourier transforms of the spin field with the smallest possible wavevectors, which are oriented in the various lattice directions, or a parameter explicitly characterizing the interface position, as defined elsewhere (Wang *et al.*, 1989). In addition, to the structure function and direct microscopy, the electric polarization and associated susceptibility should help characterize the change of order at the transition temperature.

The essential anisotropy of the phase segregation in this problem appears to compel one to consider separately the values of the relevant quantities along the two principal directions of the lattice, as it is in fact suggested by definition (3.17). Such a detailed

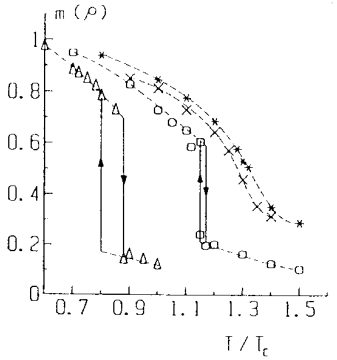


Figure 7 The order parameter (3.17) for the system in Figure 5, as a function of temperature for different values of the density, $n = \frac{1}{2}$ (asterisks), 0.35 (crosses), 0.2 (circles), and 0.1 (triangles). The arrows represent transitions observed during the Monte Carlo experiment between the two branches obtained when $n \leq 0.2$ as the system is heated up from zero temperature or cooled down from infinite temperature, respectively.

study (Vallés and Marro, 1987), however, revealed no noticeable results for the purposes of this paper. Consequently, we shall be mainly concerned with global quantities such as the configurational energy, defined as $e(T) \equiv -\langle H \rangle_T / JN = 2 - 2u(T)$, and the associated specific heat C, defined as the temperature derivative of e or u (after the appropriate smoothing of the data by spline interpolation). Figure 8 depicts the behavior of C. This confirms in particular a sharp discontinuity for $n \leq 0.2$ and an acute peak for $n = \frac{1}{2}$ while the case $n = 0.35$ is ambiguous: it may be that the discontinuity there is too weak to be evidenced by the present MC data. Most interesting also is the comparison between the behavior of C defined as the derivative of the energy and that of the mean squared fluctuations of the energy data (and a similar comparison concerning the order parameter susceptibility). It then follows no general confirmation of the fluctuation-dissipation theorem for the present non-equilibrium situation. Actually, there is no general proof here of the microscopic reversibility leading to that theorem since the detailed balance condition only holds locally in the model, and the external electric field energy cannot be included in the Hamiltonian.

The particle current in the direction of the field, j_x, i.e. the number of actual jumps per site performed in the direction $\hat{\mathbf{x}}$ per unit time, for the infinite lattice is represented in Figure 9. This reveals itself qualititatively similar to some related measurements in real FIC materials. Let us study the experimental situation more closely. It is usually credited (e.g. the case of α-AgI) that a type of ions (Ag^+) move in a liquid-like fashion through a solid lattice set up by another type of ions (I^-), thus producing

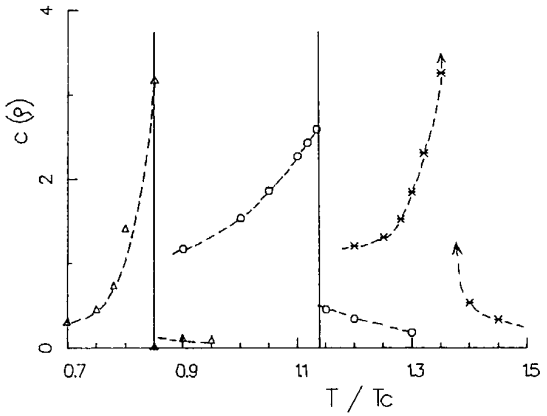
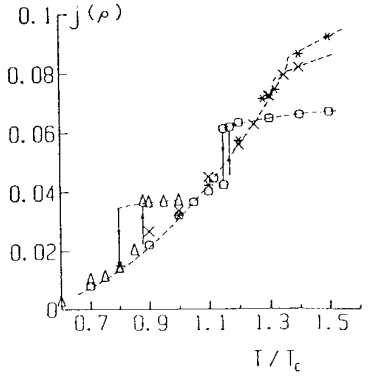
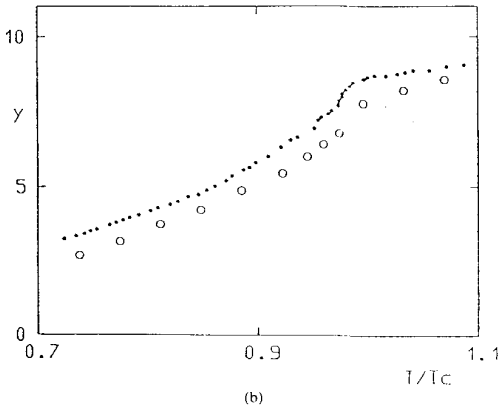


Figure 8 Variation with temperature and density of the nonequilibrium "specific heat", obtained as the temperature derivative of the configurational energy for a lattice $L \times L$: $n = 0.1$ (triangels, $L = 100$), $n = 0.2$ (circles, $L = 100$), and $n = \frac{1}{2}$ (asterisk, for $L = \infty$ as obtained from a finite size scaling analysis). The lines are a guide to the eye.



(a)



(b)

Figure 9 The electric or particle current in the direction of the field for the system in Figure 5, as a function of temperature and particle density: (a) for $L = 100$ and different values of n, using same symbols as in Figure 7, and (b) for $L \to \infty$ and $n = \frac{1}{2}$ (empty circles, $y = 12.2 j_x$) and for $AgCrS_2$ (Hibma, 1980) (asterisk, $y = 3(\sigma T_c)^{0.2}$ where σ represents the ionic conductivity in units of $(\Omega\text{-cm})^{-1}$ and $T_c = 673$ K); i.e. the units for the vertical axis in the latter are arbitrary.

ionic conductivities of the order of 1 Ω^{-1} cm^{-1} below the melting point (as compared to 10^{-8} Ω^{-1} cm^{-1} for the solid NaCl at 200°C). Nevertheless, while the onset of the high conductivity phase is usually recognized as devising a typical phase transition phenomenon, the reported nature of those phenomena is rather varied and, sometimes, puzzling. As a good starting point, one may refer to the classification of Boyce and Huberman (1979). This sketches types I, II and III of FIC materials whose main features may be well understood at the light of the behavior of the present model:

Type I is characterized by an obvious discontinuity at the transition temperture, e.g. the case of AgI which presents a four decades jump in the conductivity. Even though the experimental results are not very clear-cut at present, e.g. due to the presence of polymorphic transitions and other effects, the situation seems typically that of a first-order phase-transition, and it is well mimetized by the model case with $n \ll \frac{1}{2}$. As a matter of fact, the outstanding mobility of the conducting ions, whose number is typically of the order of the Avogadro number, is only conceivable in the presence of a large amount of vacant positions in the lattice (either lattice or interstitial sites). In some cases, large mobilities will be due to the "melting" of a sublattice at the transition temperature, thus inducing a large degree of disorder which should be detectable. In fact, a change of entropy which is comparable to the one for the melting in the corresponding material was measured sometimes (O'Keeffe and Hyde, 1976; Hayes 1978). In other cases, the high conductivity will be allowed for by a large concentration of ionic defects. The latter seems the case of ice where it was recently reported a first-order phase transition at high temperature (Ryzhkin, 1985).

Type II is characterized by a more continuous behavior, i.e. the corresponding materials show a continuous conductivity-temperature curve with a change of slope at the transition temperature. Under ideal conditions, that change should probably be expected to be rather abrupt, as it was reported for $AgCrS_2$; chances are that the gradual change observed in PbF_2 is rather associated to a complex experimental situation, e.g. one producing a "diffuse transition". One is tempted to identify type II with the occurrence of a second-order phase transition, as in the model case with $n = \frac{1}{2}$. Nevertheless, we believe that the materials of type II might rather undergo a weak first-order transition [see, for instance, Gurevich and Kharkats 1986] or a transient situation such as the one in the model for $n = 0.35$. In fact, as it was revealed above by the behavior of the model, one needs to perform a detailed analysis of specific heat and other quantities, besides order parameter and electric current, before safely determining the order of the involved phase transition. It might also be very helpful to investigate the structure function and the occurrence of transient or metastable states, as discussed below.

It was also reported the existence of a Type III of materials where the conductivity apparently increases exponentially over a broad range of temperatures. Assuming good, ideal experimental conditions, such a behavior seems to reflect the absence of actual long-range order. For instance, a situation similar to the one in the one-dimensional version of the model in Section 3.1 under a mean-field treatment. Actually, it seems that any classification of FIC should pay attention first to the dimension of the space where the ionic conductivity predominantly occurs.

The MC values for m may be analyzed by finite size scaling techniques and other means (Vallés and Marro, 1987) to obtain transition temperatures (cf. Table 1) and, for $n = \frac{1}{2}$, the relevant critical behavior for the infinite system. It thus follows in particular that $T_c^\infty = (1.355 \pm 0.003)T_c^0$, where $T_c^0 \approx 2.2692\ J/k_B$, and that the corresponding critical exponent is $\beta = 0.230 \pm 0.003$. That is, the present nonequilibrium system has a critical behavior differing from the familiar ones, namely from the Onsager equilibrium one ($\beta = \frac{1}{8}$) and from the Landau classical one ($\beta = \frac{1}{2}$); actually, a more recent study demonstrates $\beta < 0.5$ and confirms $\beta \approx 0.23$ for the present model [Marro *et al.* 1989]. It should be noted that a related model with competing dynamics under the action of an external field was recently also found to be characterized by $\beta \approx 0.22$ [Wang *et al.*, 1989], and we found that this also seems the value characterizing a two-dimensional Ising model where the transitions along the $\hat{\mathbf{x}}$ direction are performed "as if the bath temperature was infinite" and those perpendicular to $\hat{\mathbf{x}}$ occur at a finite temperature T. In a sense, this is amazing since it was argued for some time that nonequilibrium systems might be characterized by classical behavior, as seems to occur in some significant cases (see, for instance, Onuki and Kawasaki, 1981; see also van Beijeren and Schulman, 1984; Krug *et al.*, 1986; Wang and Lebowitz, 1988). It is not surprising however that nonequilibrium critical behavior is more complex and varied than the equilibrium one, given the singular simplicity of the equilibrium state. It thus seems that the electric field is a relevant parameter of the model, in the sense that it may essentially modify the critical behavior of the system with $E = 0$. To the best of our knowledge, there is no detailed study of critical properties of FIC (real) materials under nonequilibrium conditions reporting on the relevance of the parameter E; see, however, Gurevich and Kharkats (1986), Pardee and Mahan (1974), and Vargas (1978) suggesting critical behaviors close to the one for the equilibrium Ising model. Consequently, more experiments in this area would be very helpful, specially those concerning low-dimensional materials and "large" (saturating) external electric fields focusing on the nature of the nonequilibrium phase transition and critical properties.

The study of the variation with system size of the order parameter and other quantities is also interesting (Vallés and Marro, 1987; Wang *et al.*, 1989). The main

Table 1 Values for the tranistion temperature as a function of n for the two dimensional system obtained by Monte Carlo methods. The error bars represent the location of the closest metastable states observed during the system evolution.

n	T_c^∞
0.50	1.355 ± 0.003
0.35	1.32 ± 0.02
0.20	1.16 ± 0.02
0.10	0.84 ± 0.04
0.075	0.67 ± 0.13
0.05	0.37 ± 0.18

qualitative conclusion is that m has strong finite-size effects both above and below T_c^∞ and, in particular, that the situation here rather resembles that found in the equilibrium case with free edges (Landau, 1976). The specific scaling study of the present anisotropic situation confirms the above critical behavior and it also indicates that $\nu \approx 0.55$–1 and $\alpha = 0$ or very small.

MC simulations have also monitored the time evolution, e.g. from initial random configurations, as if the system was quenched from an "infinite" temperature. In the relaxation of systems with $0.35 \leq n \leq \frac{1}{2}$ below the coexistence curve, we often observed that they first evolve towards intermediate states characterized by several strips which are parallel to the field direction; cf. Figure 6. On some occasions, these suddently decayed into one-strip states after rather large evolutions, while the system was never seen to escape from those multi-strip states at low enough temperatures. Again, this is a size dependent effect: the number of strips increases with L, and the corresponding time of escape seems to diverge as $L \to \infty$. The situation differs however for $n \leq 0.2$, where there may not be enough particles in the system to build up those intermediate states, and the evolution most times proceeds directly through one-strip states which become more and more compact with time.

In addition to those intermediate multi-strip states, the system with $n < \frac{1}{2}$, and more evidently for $n \leq 0.2$, presents well defined metastable states. The former may only be termed metastable in the sense that they fulfil a necessary criterion (Binder and Müller-Krumbhaar, 1974). That is, one may define a function $\Phi_A = [A(t) - A(\infty)]/[A(0) - A(\infty)]$ associated to any relaxing macroscopic quantity A; a metastable state may then be characterized by the absence of any evolution in a time scale which is macroscopically large enough and, consequently, by the presence of a plateau in the function $\Phi_A(t)$. This is certainly the case for multi-strip states when A represents, for instance, the order parameter or the configurational energy. Actually, the life time of those intermediate states increases with decreasing temperature and it seems to diverge in some cases for $n = \frac{1}{2}$. Nevertheless, multi-strip states are not true metastables according to the three properties listed by Penrose and Lebowitz (1971) for characterizing metastable states: (i) only one thermodynamic phase is present (and usual thermodynamics applies to it); (ii) an (isolated) system in a metastable state is likely to take a very long time, possibly years, to get out; and (iii) escape from the metastable state is an irreversible process. The multi-strip states fulfil (ii), as noticed before, and (iii), in fact reversibility for $n \leq \frac{1}{2}$ was never observed. However, they are segregated states violating (i). What is more, their nature is size dependent, and $\Phi_m(t)$ evolves in those cases via several abrupt steps. By contrast, the metastable states reported above for $n < \frac{1}{2}$ satisfy the three criteria. In any case, it may be noticed that both, multistrip and real metastable states are here a non-equilibrium effect, and that the former do not seem to have an equilibrium counterpart.

3.3 *Monte Carlo results for d = 3*

As mentioned before, the conductivity in FIC materials frequently occurs as a low dimensionality effect. Consequently, one- and two-dimensional models have an unusual physical relevance, on the one hand, and it results interesting the study of the model properties as one varies d, on the other. We shall briefly describe some unpublished MC results for $d = 3$ which, in particular, confirm some of our previous statements for $d < 3$. The data refer now to $L \times L \times L$ s. c. lattices, mostly $L = 30$, with $n = \frac{1}{2}$ and periodic (toroidal) boundary conditions, evolving under different dynamics for an "infinite" external electric field and $\Gamma = 1$. The rates used here by us are (2.5a), (2.5b), and a generalization of the dynamics in the model by van Beijeren and Schulman (1984), namely $c = 1$ for jumps in the direction of the field, $+\hat{\mathbf{x}}$, $c = 0$ in the opposite direction, $-\hat{\mathbf{x}}$, and $c = \exp(-\delta H/2k_BT)$ for jumps perpendicular to the field.

It happens that the data, e.g. the order parameter and the electric current, obtained for different rates may be scaled together by simply renormalizing units appropriately; see Figure 10. This suggests universality with respect to dynamics. That is, even though the corresponding critical temperatures differ from each other, namely $T_c^\infty/T_c^0 = 1.14$, 1.19 and 1.35 (± 0.02) respectively for the rates mentioned above, the

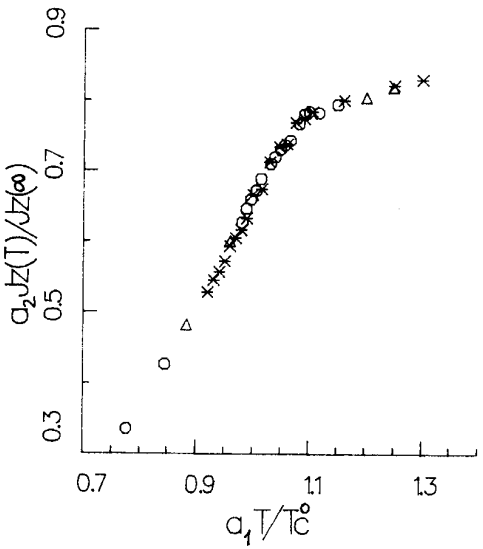


Figure 10 Same as Figure 9, normalized to the infinite temperature value $J_z(\infty)$, for the three-dimensional model as obtained from MC simulations for $n = \frac{1}{2}$ and various dynamics, namely for (2.5a) (asterisks), for (2.5b) (triangles), and for the dynamics refined in section 3.3 (circles). The temperature is in units of T_c^0/a_1; $a_1 = 1$, 0.96, 0.845 and $a_2 = 1$, 0.99, 0.96 for the three cases respectively. The system energy has a similar behavior.

data is always consistent with the same value for the critical index, apparently $\beta = 0.4 \pm 0.1$, independent of dynamics. Incidentally, this seems to confirm that the model belongs to a universality class differing from both the classical mean-field ($\beta = \frac{1}{2}$) and the equilibrium Ising ($\beta \approx 0.313$) ones. Unfortunately, the latter values of β are too close to each other (for $d = 3$) to allow a definite conclusion in that sense, given the reported error bars of the data at hand. Much more MC computation will be needed before fully determining the critical behavior of the three-dimensional model. That task has revealed itself as far from easy, however, due to a significant degeneracy of the steady state, i.e. the system shows up ordered at low temperatures forming nothing as simple as a strip but a more complexd anisotropic structure. It also deserves a comment the fact that the critical temperature may be obtained from the behavior of the order parameter and also, as expected, from both, a well-defined change in the slope of the current-temperature curves (cf. Figure 10), and a sudden, large increase in the maximum value of the structure function (e.g. after performing a circular average) as one raises the temperature.

3.4 Repulsive interactions

Some pioneering MC work on this problem (Katz *et al.*, 1983) revealed that, while the critical temperature induced by the field is finite and larger than T_c^0 for $J > 0$, when $J < 0$ the field lowers the transition temperature and the state becomes disordered for strong enough fields. This is puzzling because some real materials reported as being of types I or II, i.e. systems presumably undergoing first- or second-order phase transitions, apparently contain antiferromagnetic couplings. One may argue, given the evidence above, that those materials instead have predominant or effective ferromagnetic-like interactions. That belief could be corroborated by experiments, e.g. by studying the (nonequilibrium) critical behavior of FIC materials. In fact, recent Monte Carlo simulations for $J < 0$, $n = \frac{1}{2}$, and dynamics (2.5a), supported by a field-theoretic model, suggests that the field parameter E is then irrelevant for the critical behavior, e.g. in the sense that the second-order phase transitions when $J < 0$ are characterized by $\beta = \frac{1}{8}$ for $d = 2$ (Leung *et al.*, 1989).

More precisely, that study (Leung *et al.*, 1989) (see also a related mean-field computation by Dickman (1990)) allows one to outline the phase diagram in a corner of the (E, T) plane, actually for $\frac{1}{2}J < E < 2.5J$ and $0.2T_c^0 < T < 1.2T_c^0$. It follows the existence of a line of second-order transitions, starting at $(0, T_c^0)$, which seems characterized at $d = 2$ by the Onsager equilibrium critical exponents. Indeed, it is observed for $E = \frac{1}{2}J$ a clear second-order transition with $T_c(E) = (0.81 \pm 0.01)T_c^0$, $\beta = \frac{1}{8}$ and $\nu = 1$. There is also a line of first-order transitions emerging from the $T = 0$ axis for larger values of E, the two lines probably joining at a (nonequilibrium) tricritical point located around $E \approx J$ and $T \approx T_c^0$. That is, the situation here resembles more the one in other nonequilibrium models (Garrido *et al.*, 1989, 1990a; Lebowitz *et al.*, 1988) than the case $J > 0$ above. Moreover, when the field is increased further than E_c ($E_c = 2J$ for $T > 0$, and $E_c = 3J$ for $T = 0$), it overcomes the repulsive force and long-range order is destroyed for any T, in contrast to the attractive case; this probably occurs also for any deviation from $n \equiv \frac{1}{2}$. The antiferromagnetic case also differs from the case $J > 0$ in that anisotropic effects are mostly negligeable and small even in the critical region. Summing up, even though the model with $J < 0$ has a very rich behavior deserving further theoretical study, it seems to have a lesser practical relevance than the case $J > 0$ in relation to the behavior of FIC materials.

4 HYDRODYNAMICS

The old problem of deriving macroscopic hydrodynamic-like equations from microscopic models of interacting particles has recently benefited from a series of successful efforts (De Masi *et al.*, 1986; Lebowitz *et al.*, 1988; Fritz and Maes, 1988). In order to illustrate both that approach and the versatility of the model in this paper in producing relevant macroscopic information, we sketch here an original derivation of a specific hydrodynamic equation concerning FIC behavior. A similar equation for a related Ginzburg–Landau model has been obtained by Fritz and Maes (1988).

The Master Equation (2.2) determining time evolution may be written for $E = 0$ as $dP(\sigma; t)/dt = LP(\sigma; t)$ where L represents the operator defined by the r.h.s. of Eq. (2.2) (after setting $E = 0$ there). That is, L represents the action of a transition probability per unit time satisfying the condition of detailed balance: $c(\sigma^{x,y}; \mathbf{x}, \mathbf{y}) \exp[-\beta H(\sigma^{x,y})] = c(\sigma; \mathbf{x}, \mathbf{y}) \exp[-\beta H(\sigma)]$. Hence, the choice $c(\sigma; x, y) = \Phi_0(\beta\delta H)$ implies $\Phi_0(h) = \Phi_0(-h)\exp(-h)$ as it occurs with (2.5). The field may be introduced as a perturbation, i.e.

$$dP_E(\sigma; t)/dt = LP_E(\sigma; t) + \varepsilon L_E P_E(\sigma; t), \tag{4.1}$$

where L_E has precisely the structure of the r.h.s. of Eq. (2.2), $L_{E=0} = L$, and one defines Φ_E similarly. When $E = 0$, both (2.2) and (4.1) have the same (equilibrium) solution, (2.3); when $E \neq 0$, (2.2) and (4.1) only differ in that the latter involves an additional (trivial) canonical mechanism.

To obtain a hydrodynamic description from (4.1), one needs to go from the microscopic lattice to the macroscopic continuum and, simultaneously, to describe the system on a time scale where the effects of individual, microscopic events become negligible. With that aim, one scales lengths according to $r = \epsilon x$ and times according to $\tau = \epsilon^2 t$, and considers the limit $\varepsilon \to 0$, $x \to \infty$, $t \to \infty$, with r and τ remaining finite. Under those conditions, the effect of the field becomes *microscopically* negligible as compared to the canonical dynamics, while it remains *macroscopically* measurable. Actually, the system then approaches a sort of local equilibrium (De Masi *et al.*, 1984; Garrido *et al.*, 1989) with respect to the equilibrium distribution (2.3). Consequently, under that limit $\langle \sigma_x \rangle_t \equiv \sum_\sigma \sigma_x P_E(\sigma; t)$ is transformed to the deterministic variable $m(r; \tau)$ and, when one considers the one-dimensional system, the latter satisfies, as implied by (4.1), the hydrodynamic equation:

$$dm(r; \tau)/d\tau + dj(r; \tau)/dr = 0 \tag{4.2a}$$

where

$$j(r; \tau) = j_0(r; \tau) + j_E(r; \tau). \tag{4.2b}$$

Here,

$$j_0(r;\tau) = -(dm/dt)\{1 + \tfrac{1}{2}[1 - m(d\alpha/dm) + \alpha][\Phi_0(k) - \Phi_0(0)]\}, \quad (4.2c)$$

$k \equiv 4\beta J$, and

$$j_E(r;\tau) = \tfrac{1}{2}m^2[\Phi_E(k-E) - \Phi_E(k+E)] - \tfrac{1}{4}\alpha^2[\Phi_E(E) - \Phi_E(-E)] + \tfrac{1}{4}(\alpha + \tfrac{1}{2}\alpha^2)[\Phi_E(k+E) + \Phi_E(-k-E) - \Phi_E(k-E) - \Phi_E(-k+E)] \quad (4.2d)$$

where $\alpha = \alpha(m) \equiv [e^{-(1/2)k}(2m^2 - 1) + \eta][e^{-(1/2)k} + \eta]^{-1}$, $\eta \equiv \{1 - m^2[1 - e^{-k}]\}^{1/2}$, and $m = m(r;\tau)$. Several interesting facts follow from Eq. (4.2), e.g. expressions for the coefficient of bulk diffusion,

$$D(m) = 1 + (1 - m^2)[\Phi_0(k) - 1]\{1 - m^2[1 - e^{-k}] + e^{-(1/2)k}\}^{-1}, \quad (4.3)$$

and for the conductivity,

$$s(m) = -(1 - m^2)\varphi_+ + \tfrac{1}{2}(1 - \alpha)[\varphi_+ - \varphi_-] + \tfrac{1}{4}(1 - \alpha^2)[\varphi_+ + \varphi_- + 1], \quad (4.4)$$

where $\varphi_\pm \equiv [d\Phi_E(u)/du]_{u=\pm k}$. Thus, $m = 0$ leads to the (macroscopic) Einstein relation $s(0) = D(0)[1 + e^{-2\beta J}]^{-1}$ for any microscopic dynamics such that $\Phi_0 \equiv \Phi_E$, and also, for instance, to the predictions that

$$D(0)_{J>0} = \exp(2\beta|J|)D(0)_{J<0} \quad (4.5)$$

and

$$s(0)_{J>0} = \exp(2\beta|J|)[1 + \exp(2\beta|J|)][1 + \exp(2\beta|J|)]^{-1} \quad (4.6)$$

for $d = 1$. The latter may be compared with a mean-field prediction in Section 3.1. Finally, another interesting fact is that Eq. (4.2) transforms under the limit $\beta = 0$, $E \to \infty$ into the familiar Burgers equation for a "simple asymmetric exclusion process" [Burgers 1974), i.e.,

$$dm/d\tau + K(dm^2/dr) = d^2m/dr^2, \quad (4.7)$$

where $K \equiv \frac{1}{2}[\Phi_E(-\infty)^- \Phi_E(\infty)]$, and for a field large enough it follows the useful approximate result (see, for instance, Martin *et al.*, 1978; De Dominicis and Peliti, 1978) $j_E(r;\tau) \approx Km(r;\tau)$.

5 CONCLUSIONS

Contrary to the situation which is now rather familiar in equilibrium, relatively complex nonequilibrium macroscopic phenomena can seldom be formulated microscopically. We discussed here one of those rare cases: The observed behavior of fast (or super) ionic conductors (FIC) may sensibly be understood, even though the model involves a remarkable oversimplification of Nature, by means of lattice-gas models which are driven to (nonequilibrium) steady states by an external electric field E. Thus, at the light of a comparison between the macroscopic behavior of the model and available observation on real systems, we were able to conclude both expected macroscopic features of FIC materials and basic microscopic ingredients determining that behavior.

For instance, it follows that some properties may depend crucially on the dimension of the space where the ionic conductivity predominantly occurs. When that is mainly restricted to channels, the model suggests the occurrence of two qualitatively different behaviors. A system with $J < 0$ (short-ranged repulsive interactions) and $n \neq \frac{1}{2}$, is expected to present similar good conductor states both at low and high temperatures, with a sudden current increase at some temperature (around $T = 4/E$ in the model). On the contrary, for $J < 0$ (attractive interactions between particles) and $n = \frac{1}{2}$ (half-filled lattice), or $J > 0$ at any density, one can expect a continuous crossover from an insulator state when $T \ll 4/E$ to a good conductor state when $T > 4/E$, the smooth transition being located, for instance, by a maximum of the specific heat which increases with E. None of those behaviors is to be associated with the onset of long-ranged order, i.e. one does not expect to have a phase transition at finite temperature for $d = 1$, unless mean-field effects are as predominant in real materials as they are in the $\Gamma \to \infty$ version of the model, a situation which is not supported by the analysis here. Moreover, given that those differences are observable in principle, they could help to determine the sign of the effective interionic interactions in one-dimensional FIC materials. Also interesting is the prediction that, for a given temperature and strength of interactions, the system conductivity is larger for $J < 0$ than for $J > 0$. On the other hand, we presented the derivation of a hydrodynamic equation which leads in particular to expressions for transport coefficients such as conductivity and bulk diffusion.

Two-dimensional systems with $J > 0$ are expected to undergo phase separation at low temperatures, with the transition temperature $T_c(E)$ an increasing function of E which has an upper limit $T_c^\infty > T_c^0$ (T_c^0 is the equilibrium, Onsager critical temperature). More precisely, the model under saturating fields has a critical point at density $n_c = \frac{1}{2}$ which is characterized by novel critical exponents (e.g. $\beta \approx 0.23$), it undergoes first-order transitions for $n \ll n_c$, and the discontinuities, if any, are extremely weak when $n_c > n > 0.35$. The segregation is always highly anisotropic. The system with $n \approx n_c$ relaxes at low temperatures into a particle-rich strip along the field direction which coexists with a homogeneous particle-poor gas. However, the system with $n_c < n \leq 0.35$ first segregates into intermediate multi-strip states, the time of escape increasing with decreasing temperature, and it also presents real, non-segregated metastable states, more evident as the density is lowered. Thus, one may outline coexistence and spinodal lines. The former is halfway between the equilibrium one by Onsager and the classical one by Landau, the latter is a mean-field feature but it lies closer to $T_c(n)$ than predicted by classical theory.

The model computation of several physical quantities (order parameter, energy, "specific heats", structure function, ordering susceptibility and electric current) for $d = 2$ permits to make contact with experimental observations. For instance, it helps the interpretation of the reported existence of two types of FIC materials: Type I would correspond to the case $n \leq 0.2$ in the model, while the materials termed Type II seem to include both the case of a critical density and the case $n_c > n > 0.35$. Computations for a fully three-dimensional system with $J > 0$, though less definite at present, seem consistent with the situation for $d = 2$.

It is more difficult to assert the relevance of the model with $J < 0$ for understanding

FIC behavior. For $n = \frac{1}{2}$ and $J < 0$ the model predicts second-order phase transitions with equilibrium exponents for small fields, first-order ones for intermediate fields, and lack of order at any temperature for large fields and also for any $n \neq \frac{1}{2}$. We found no experimental evidence supporting that picture, so that we argue that either most FIC materials have predominant or effective (short-ranged) attractive interactions between particles, or else that the hard-cored interaction which incorporates the model with $J > 0$ is the only repulsive attribute needed to obtain FIC behavior.

Finally, it may be stressed that the model for $d = 2$ (perhaps also for $d = 3$) strongly suggests in particular the existence of a new universality class which may accommodate many FIC materials. This is characterized by *nn* attractive interactions besides a hardcore repulsion, by the existence of an external agent influencing correlations along one of the system directions, and by a set of values for the critical indexes differing from the familiar ones; similar values were recently found in two more related nonequilibrium models. New experimental measurements on FIC materials with effective dimensionality $d = 1$, 2 or 3 under nonequilibrium conditions would greatly help the present understanding.

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