

Monte Carlo Study of the Generalized Reaction-Diffusion Lattice-Gas Model System

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The reaction-diffusion lattice-gas model is an interacting particle system out of equilibrium whose microscopic dynamics is a combination of Glauber (reaction) and Kawasaki (diffusion) processes; the Glauber rate $c(\mathbf{s}; \mathbf{x})$ at site \mathbf{x} when the configuration is \mathbf{s} satisfies detailed balance at temperature T , while the Kawasaki rate $\Gamma c(\mathbf{s}; \mathbf{x}, \mathbf{y})$ between nearest-neighbor sites \mathbf{x} and \mathbf{y} satisfies detailed balance at a different temperature T' . We report on the phase diagram of that system as obtained from a series of Monte Carlo simulations of steady states in two-dimensional lattices with arbitrary values for T' , T , and Γ ; this generalizes previous analytical and numerical studies for $\Gamma \rightarrow \infty$ and/or $T' \rightarrow \infty$. When the rates are implemented by the Metropolis algorithm, the system is observed to undergo various types of first- and second-order (nonequilibrium) phase transitions, e.g., one may identify Onsager (equilibrium) as well as Landau (mean-field) types of continuous phase transitions.

KEY WORDS: Nonequilibrium steady states; reaction-diffusion stochastic models; competing dynamics; nonequilibrium phase transitions; Monte Carlo simulations.

Consider a lattice-gas model system on a square lattice with $N = L^2$ sites. The configuration $\mathbf{s} = \{s_x; \mathbf{x} \in \mathbf{Z}^d, s_x = \pm 1\}$ evolves in time due to a combination of two independent processes. Namely, \mathbf{s} changes stochastically due to both Glauber⁽¹⁾ creation-annihilation or *reaction* processes governed by a heat bath at temperature T , and Kawasaki⁽²⁾ *diffusion* processes caused by nearest-neighbor (nn) exchanges which occur as if the associated bath temperature was T' instead. Consequently, the configurational

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probability distribution $\mu(\mathbf{s}; t)$ evolves in time according to a Markovian master equation⁽³⁾:

$$d\mu(\mathbf{s}; t)/dt = (\mathbf{L}_G + \Gamma\mathbf{L}_K) \mu(\mathbf{s}; t) \quad (1)$$

where Γ is the ratio of attempted exchanges per bond to attempted reactions per site, $\mathbf{L}_G \equiv \sum_x [\mathbf{G}_x - 1] c(\mathbf{s}; \mathbf{x})$, with $\mathbf{G}_x g(\mathbf{s}) = g(\mathbf{s}^x)$, $\mathbf{L}_K \equiv \sum_{|\mathbf{x} - \mathbf{y}|=1} [\mathbf{K}_{\mathbf{xy}} - 1] c(\mathbf{s}; \mathbf{x}, \mathbf{y})$ with $\mathbf{K}_{\mathbf{xy}} g(\mathbf{s}) = g(\mathbf{s}^{\mathbf{xy}})$, and $g(\mathbf{s})$ stands for an arbitrary function of the configuration. Here, \mathbf{s}^x represents the configuration obtained from \mathbf{s} by applying the operator \mathbf{L}_G , i.e., by changing s_x to $-s_x$, and $\mathbf{s}^{\mathbf{xy}}$ is the one obtained from \mathbf{s} after the interchange of the occupation variables at sites \mathbf{x} and \mathbf{y} as performed by \mathbf{L}_K .

The respective rates $c(\mathbf{s}; \mathbf{x})$ and $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$ for the reaction and diffusion processes both satisfy detailed balance, but with respect to different heat temperatures. That is,

$$\mathbf{L}_G \mu_{\text{eq}}^T(\mathbf{s}) = 0 \quad (2)$$

with

$$\mu_{\text{eq}}^T(\mathbf{s}) = \left\{ \sum_{\mathbf{s}} \exp[-H(\mathbf{s})/k_B T] \right\}^{-1} \exp[-H(\mathbf{s})/k_B T] \quad (3)$$

where $H(\mathbf{s})$ represents the system configurational Hamiltonian, which will be assumed for simplicity to be

$$H(\mathbf{s}) = -J \sum_{|\mathbf{x} - \mathbf{y}|=1} s_x s_y, \quad J > 0 \quad (4)$$

and

$$\mathbf{L}_K \mu_{\text{eq}}^{T'}(\mathbf{s}; m) = 0 \quad (5)$$

with

$$\mu_{\text{eq}}^{T'}(\mathbf{s}; m) = Z(\hat{\mu})^{-1} \exp \left[-H(\mathbf{s})/k_B T' + \hat{\mu} \sum_{\mathbf{x}} s_x \right] \quad (6)$$

Here, $\hat{\mu} = \hat{\mu}(m)$, namely $m = N^{-1}(d/d\hat{\mu}) \ln Z(\hat{\mu})$, where $Z(\hat{\mu}) \equiv \sum_{\mathbf{s}} \exp[-H(\mathbf{s})/k_B T' + \hat{\mu} \sum_{\mathbf{x}} s_x]$.

That model has two familiar, particularly simple versions which may serve as a canonical reference for the nonequilibrium situations of interest here. Namely, when $\Gamma \equiv 0$ or $c(\mathbf{s}; \mathbf{x}, \mathbf{y}) \equiv 0$ one recovers the Glauber case⁽¹⁾ with a nonconserved order parameter. Any rate $c(\mathbf{s}; \mathbf{x})$ satisfying detailed balance [which implies (2)] drives the system to the same steady state.

This is the equilibrium Gibbs state (3) corresponding to temperature T and energy $H(\mathbf{s})$, which is known⁽⁴⁾ to undergo, in the infinite-volume limit ($N \rightarrow \infty$), a second-order phase transition at a finite temperature T^0 . That transition also occurs (at T'^0) when $c(\mathbf{s}; \mathbf{x}) \equiv 0$ for any rate $\Gamma c(\mathbf{s}; \mathbf{x}, \mathbf{y})$ satisfying detailed balance [which implies (5)] and any finite Γ ; the system reduces now to the lattice-gas model with Kawasaki dynamics⁽²⁾ whose steady states are Gibbs states (6) at temperature T' with a fixed value m for the order parameter.

The situation is very different otherwise. That is, the steady state implied by the competing dynamics in Eq. (1) is a nonequilibrium one in general, and it may not be unique. For instance, it may depend, even qualitatively, on the specific form assumed for the reaction and diffusion rates, $c(\mathbf{s}; \mathbf{x})$ and $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$. Moreover, the infinite-volume system may suffer, as in an equilibrium condition, instabilities leading to (nonequilibrium) continuous or discontinuous phase transitions, and one should expect these and other features of steady states to depend, in some way to be determined, on the values for the parameters T , T' , and Γ and on the details of the dynamical processes.

These and other facts were already indicated by De Masi *et al.*,⁽³⁾ who considered the model in the limit $\Gamma \rightarrow \infty$ and $T' = \infty$, i.e., the most random and microscopically fastest diffusion process, as a continuation of previous interest on reaction-diffusion stochastic models.⁽⁵⁻⁸⁾ Subsequently, several versions of the same model in a d -dimensional space have been studied by different methods.^{(9-14),3} In particular, concerning the study of steady states for $d = 2$, which is our goal in the present note, they have been investigated by mean-field⁽⁹⁾ and Monte Carlo methods^(11,13) for arbitrary Γ when $T' = \infty$. The main conclusion from those studies concerns the presence of a (nonequilibrium) phase transition changing, as Γ is increased, from second to first order; i.e., it seems there is some kind of "tricritical point" separating those two behaviors. Nevertheless, given that only some one-dimensional versions of the model above have been amenable to an exact solution, the general understanding of the phase diagram is still, in spite of being quite interesting in itself and also as a paradigm of nonequilibrium phenomena, rather incomplete. For instance, concerning the case $d = 2$, both the behavior in the neighborhood of the supposed tricritical point and the qualitative details of the reported discontinuous transitions remain puzzling; also, any information concerning variations of T' is lacking. We present in this note a first attempt to study these questions. Namely, we investigate by Monte Carlo methods a two-dimensional system with

³ See ref. 10 for a review.

Metropolis rates when the relevant parameters (T , T' , and Γ) vary over their natural range $[0, \infty]$.

The procedure we followed is essentially the usual one in an MC experiment,^(13,15) except that after a site \mathbf{x} is chosen at random from a square lattice, then, with probability p , the variable s_x is exchanged with one of its nn \mathbf{y} according to a rate $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$ (which is computed as if the system was in contact with a heat bath at temperature T') and, with probability $1-p$, s_x is changed to $-s_x$ with a rate $c(\mathbf{s}; \mathbf{x})$ (computed as if the bath temperature was T). This corresponds, except for a renormalization of time units which is irrelevant for the steady state, to having $\Gamma = p/d(1-p)$, $0 \leq p \leq 1$.

The rates chosen here are the ones introduced by Metropolis *et al.*,⁽¹⁶⁾ i.e., $c(\mathbf{s}; \mathbf{x}, \mathbf{y}) = \min\{1, \exp(-\delta H/k_B T')\}$ and $c(\mathbf{s}; \mathbf{x}) = \min\{1, \exp(-\delta H/k_B T)\}$, where δH is the change in the configurational energy (4) which would cause the attempted move. That choice was only motivated by the fact that in practice it is most efficient when trying to simulate true (stabilized) "ferromagnetic ordering" disturbed by diffusion. As mentioned above, however, the choice of rates may prejudice the nature of the nonequilibrium steady state.

We considered different lattices sizes L^2 , $L \leq 100$, with periodic boundary conditions, attractive ($J > 0$) nn interactions, and a variety of values for the parameters T , T' , and p . Relatively long evolutions, typically between 10^4 and 10^6 MC steps (per lattice site), were required to obtain final steady states with good statistics. Once the final steady state was reached, we monitored in particular the short-range order parameter defined⁽¹⁷⁾ as $\sigma = \langle N_{++} \rangle \langle N_{--} \rangle \langle N_{+-} \rangle^{-2}$, where $\langle \cdot \rangle$ represents the MC average over configurations and N_{+-} , N_{++} , and N_{--} are, respectively, the number of three different nn pairs which may occur in the system, and also the long-range order parameter $m = N^{-1} \langle \sum_x \mathbf{s}_x \rangle$, the configurational energy $e = -(2JN)^{-1} \langle H \rangle$, and their respective fluctuations $C = N^{-1} [\langle H \rangle^2 - \langle H^2 \rangle]$ and $X = J [\langle \sum_x \mathbf{s}_x \rangle^2 - \langle (\sum_x \mathbf{s}_x)^2 \rangle]$. Notice that the precise relation between the latter and the derivatives de/dT , de/dT' , and $dm/d\mu$ is rather unclear in the present nonequilibrium situation.

The main global result concerns the nature of the phase transitions suffered by the system. We found it convenient to consider varying values of the "reaction temperature" T for selected pairs of values of the velocity and temperature of the diffusion process (p , T'). Also, the MC analysis becomes easier when one recognizes the existence of three different types of phase transitions as follows.

Type I is characterized by a continuous behavior of e and m , as in a second-order phase transition with a reaction critical temperature T_c which

is a function of both p and T' . The fluctuations C and X depict divergences at both sides of T_c , the one for C being completely symmetrical, and the short-range order parameter σ describes a peak (cf. Fig. 1) which is lopsided as well as the divergence of X , i.e., σ and X behave asymmetrically around T_c . The peak for σ becomes rather acute as N is increased. Representative cases showing that kind of behavior are, for instance, $(p, T'/T^0) = (0.95, 1), (0.8, 1), (0.6, 0.1), (0.6, 1),$ and $(0.6, 10)$, and also the case $(0.6, \infty)$, which was studied before in ref. 13; here $T^0 = 2.27 \text{ J/k}$ represents the equilibrium, Onsager critical temperature. Also noteworthy are the facts that the data for e and m can apparently be mapped onto a single curve by simply representing them versus $T/T_c(p, T')$, and that such a common curve seems to correspond, apart from the expected finite-size effects, to the exact Onsager solution for the equilibrium case; we shall

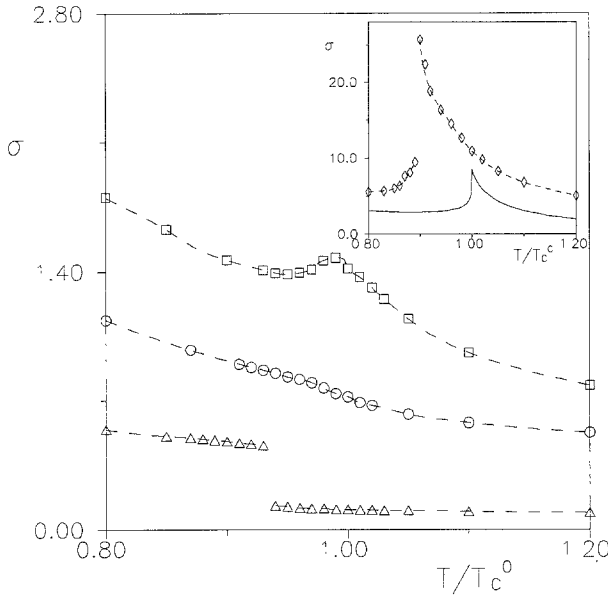


Fig. 1. The short-range order parameter σ versus reaction temperature T in units of the equilibrium, Onsager critical temperature T^0 for a given diffusion temperature, $T'/T_0 = 10$, and several velocities, $p = 0.6$ (squares, type I behavior), $p = 0.95$ (triangles, type II), and $p = 0.8$ (circles). The latter case, near the transition between types I and II that we locate around $p_i \approx 0.8$ at the indicated diffusion temperature, indicates⁽¹⁷⁾ a crossover toward classical behavior as one approaches p_i . The dashed lines are guides to the eye. The inset illustrates type III behavior (actually, the symbols are for $p = 0.95$ and $T'/T_0 = 0.1$). The solid line in the inset represents the exact equilibrium result assuming $\sigma = \frac{1}{4}[(1 + e)^2 - 4m^2](1 - e)^2$.⁽¹⁷⁾ In spite of some similarity between the two behaviors in the inset, the one obtained by MC definitely corresponds to a discontinuous phase transition with strong metastabilities.

come to this fact later. Concerning C and X , we observe a similar scaling property, except when T' considerably differs from T^0 , i.e., for $T' \gg T^0$ and for $T' \ll T^0$; such a departure from a common behavior in some occasions seems related to the expected breakdown of the fluctuation-dissipation relations. In any case, it should be noticed that, as will also come out later, the cases $T' < T^0$ and $T' > T^0$ are not equivalent, as one should probably expect.

Type II is properly represented by the cases $(p, T'/T^0) = (0.95, 10)$, $(0.95, 7)$, and $(0.85, 10)$, i.e., p is now closer to unity and $T' \gg T^0$. This is characterized (cf. Fig. 1) by well-defined discontinuities in the quantities e , m , σ , C , and X at some reaction temperature; for instance, $T_c \approx 0.955T^0$ for $(p, T'/T^0) = (0.95, 10)$. Moreover, during the time evolution of e , m , and σ near the transition point, the system usually displayed metastable states decaying toward the final steady state in a time of order 10^4 MC steps.

Finally, type III can be illustrated by $(p, T'/T^0) = (0.95, 0.1)$, $(0.95, 0.75)$, and $(0.85, 0.1)$. The system also suffers now a discontinuous phase transition, which occurs around a reaction temperature $T_c \approx 0.915T^0$ for $(p, T'/T^0) = (0.95, 0.1)$, but this essentially differs from the type II case. Actually, the respective regions for type II and type III are well separated in the phase diagram, as indicated by Fig. 3. From a practical point of view, the main difference probably concerns the fact that type III is not very suitable for an MC simulation due to the systematic presence of very slow evolutions and (long-lived) metastable states, which were typically observed to last more than 10^5 MC steps. Moreover, while the behavior of m essentially resembles the discontinuous one we described before for type II, other quantities seem to behave rather differently. The energy, for instance, while being also discontinuous at T_c , always remains larger than for type II; it is very close to the Onsager value for $T < T_c$; and it undergoes a jump which is one order of magnitude smaller than the one expected in the light of the situation for type II. There are also some obvious differences between the behaviors for σ shown by Fig. 1, and the same conclusion follows when comparing the fluctuations C and X .

We have also monitored the actual lattice configurations at selected instants of time; these greatly helped in identifying the above types of behaviors. This is illustrated by Fig. 2 confirming the existence of distinct degrees of order for the low-temperature states corresponding to systems of type II and III, respectively. That is, while the latter have the typical aspect of efficiently condensed states with long-range order which manifests itself in two well-separated phases including a rather compact liquid phase, the former are characterized by a relatively short-range order manifesting itself in a distribution of small clusters.

The relevant phase diagram of the system occurs in a three-dimen-

sional space (p, T', T) where one is interested in the reaction transition temperature T_c for each pair of values assigned to the diffusion parameters p and T' . Our results are summarized in Fig. 3. That is, for any diffusion temperature T' and $p < 0.7$, one obtains type I behavior with a critical temperature $T_c(p, T')$ which decreases monotonously with increasing T' . Increasing the value of p , however, essentially modifies the phase diagram. For instance, when $p = 0.85$ we only find type I behavior within the range $0.1 \leq T'/T^0 < 7$, while we have type II for $T' \geq 7T^0$; moreover, the

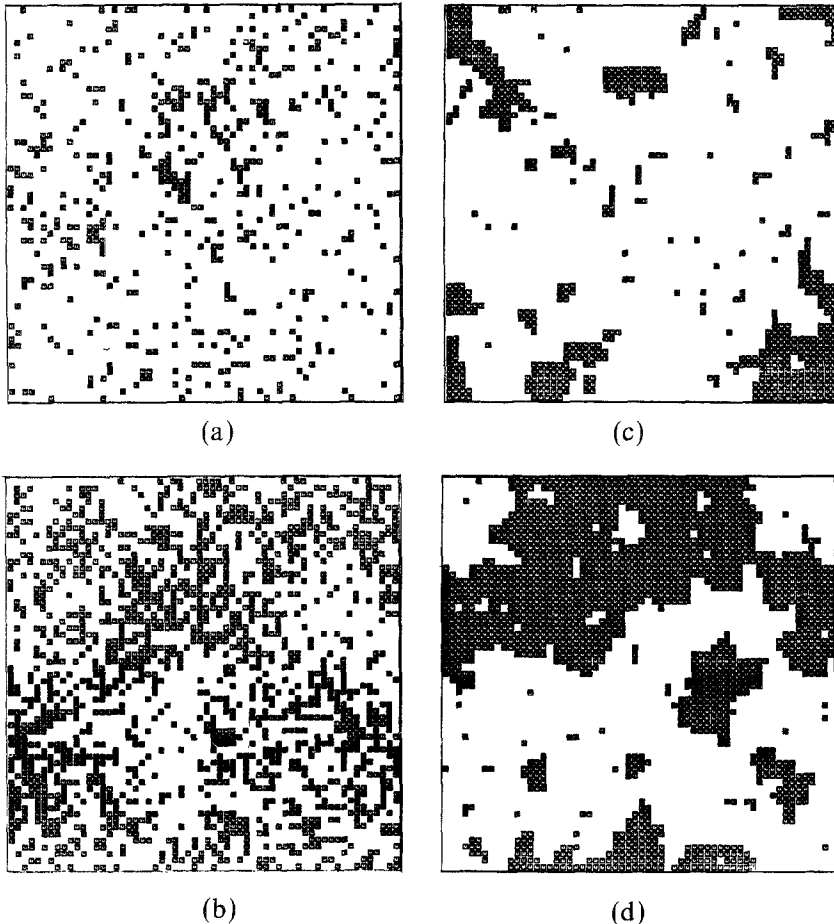


Fig. 2. Snapshots of the lattice configurations for different types of behaviors: (a) type II, namely $p = 0.95$ and $T' = 7$, where the transition occurs at $T_c \approx 0.955$, when $T = 0.95 < T_c$; (b) same, but $T = 0.96 > T_c$; (c) type III, namely $p = 0.95$ and $T' = 0.25$, where the transition occurs at $T_c \approx 0.925$, for $T = 0.92 < T_c$; (d) same, but $T = 0.93 > T_c$. Temperatures are always in units of the equilibrium critical temperature T^0 .

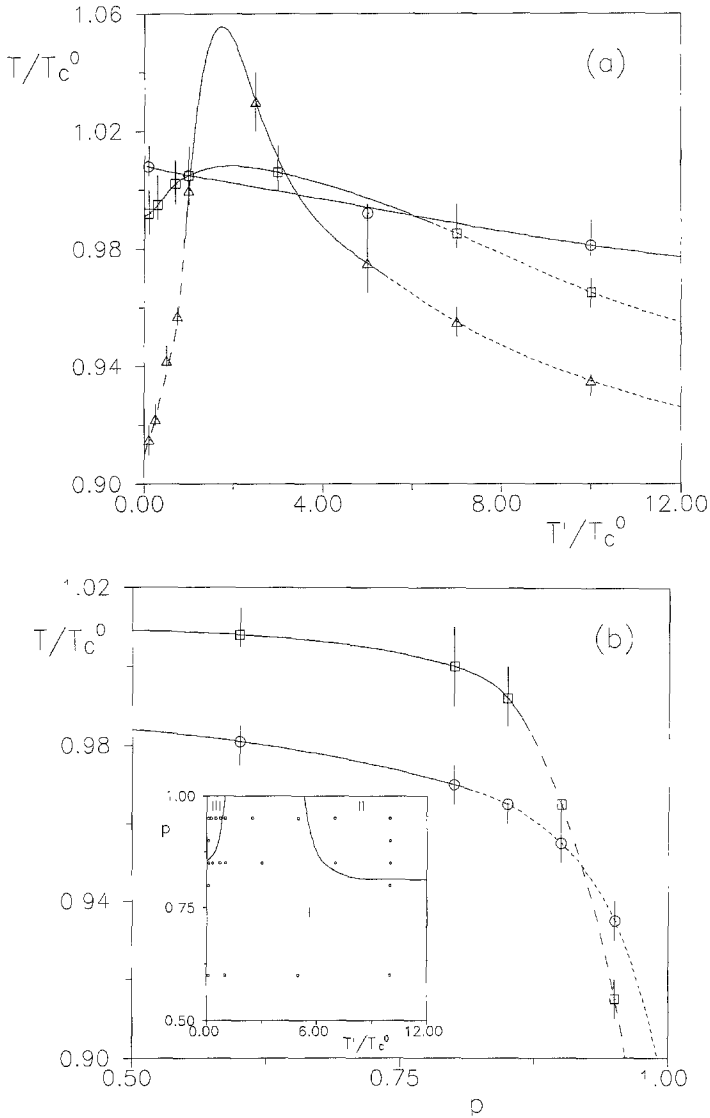


Fig. 3. Different sections of the system phase diagram as obtained from the MC analysis. The trends here are quite consistent with the exact results for $T' \rightarrow \infty$ and $p=1$ (not shown) in refs. 11 and 13. Solid lines indicate type I transitions, dotted lines are for type II, and dashed lines are for type III. The temperature is always in units of T^0 , the Onsager equilibrium temperature. The meaning of the symbols is as follows: (a) (T, T') for $p=0.6$ (circles), 0.85 (squares), and 0.95 (triangles); (b) (T, p) for $T'=0.1$ (squares) and 10 (circles). The inset represents the resulting schematic (p, T') phase diagram with the three regions of different behavior. The symbols in the inset indicate sampled points.

transition reaction temperature $T_c(p, T')$ first increases with increasing T' , reaches a maximum around $T' \approx 2T^0$, and slowly decreases afterward. Those changes occur even more markedly for $p=0.95$, where the range $0.8 < T'/T^0 < 5.5$ is characterized by type I behavior, type II occurs for $T' \geq 5.5T^0$, type III for $T' \leq 0.8T^0$, and $T_c(p, T')$ changes similarly with T' , showing a relatively steeper maximum around $T' \approx 2T^0$. It is also notable that we observe $T_c(p, T' = T^0) \approx T^0$, independent of p ; on the contrary, when one increases the diffusion velocity p for both $T'/T^0 = 10$ and 0.1 the transition temperature $T_c(p, T')$ decreases monotonously, e.g., it tends toward the exact or MC values reported in refs. 11 and 13 for $T' \rightarrow \infty$. Also, for $p > 0.8$ the transition is type I only for intermediate values of T' , while it becomes discontinuous for both small and large values of T' ; for instance, the discontinuities are type II for $T' = 10T^0$ and type III for $T' = 0.1T^0$. This situation is fully consistent with the reported existence when $T' \rightarrow \infty$ of a "tricritical point" at $p = p_t = 0.83 \pm 0.01$ which separates second-order phase transitions for $p < p_t$ from first-order transitions for $p > p_t$.⁽¹¹⁾

The MC analysis also provided some interesting information concerning the critical behavior in case of type I phase transitions. That is, most data in that region indicate that the system undergoes a continuous phase transition which is qualitatively indistinguishable from the one occurring for equilibrium states.⁽⁴⁾ More precisely, in addition to the simple scaling reported before, the present nonequilibrium phase transitions are typically characterized by Onsager critical exponents. In fact, the data in region I of Fig. 3b usually suggest that $\beta \approx 0.125$ and $\gamma' \approx 1.75$. Such a consistency between the data here and the Onsager solution, however, clearly breaks down as one approaches the region in the phase diagram corresponding to type II behavior. That is, the type I data which are close to the region corresponding to that first-order phase transition strongly suggest a changeover from Onsager to Landau⁽¹⁸⁾ (i.e., classical or mean-field) critical exponents. That changeover in the value for the critical exponents is also evidenced by the behavior of the short-range order parameter σ . That fact is illustrated by the main graph in Fig. 1, representing $\sigma(T)$ for $T' = 10T^0$ and selected values of p , namely for $p = 0.6, 0.8,$ and 0.95 (one obtains a similar situation when one fixes p and varies T'): While the graph for $p = 0.6$ (which is well inside the type I region) presents a peak whose finite-size scaling analysis⁽¹⁷⁾ makes it quite consistent with the one obtained from the Onsager solution (see the inset for Fig. 1), the one for $p = 0.8$ (which is very near the presumed transition from type I to type II regions) depicts a smooth monotonous behavior. This is indeed a confirmation of that changeover in critical behavior, because a recent study of the scaling and other general properties of $\sigma(T)$ has proved⁽¹⁷⁾ that any non-

classical critical behavior is associated with the existence of a finite peak around T_c , while the peak is absent for a continuous phase transition with classical exponents, in particular when $\beta = 1/2$ and $\alpha = 0$.

Summing up, we have studied by Monte Carlo methods a two-dimensional lattice gas or Ising model with attractive nearest-neighbor interactions whose dynamics consists of a competition between reaction (creation-annihilation or spin-flip) and diffusion (nearest-neighbor exchange) processes, with $p/2(1-p)$ the relative probability of attempted exchanges per bond to attempted flips per site, both driven by canonical heat baths characterized by different temperatures, say T and T' , respectively. This generalizes previous extensive studies by Lebowitz and collaborators^(3,10,11) and by others^(5-9,12-14) mainly concerned with the rigorous derivation of hydrodynamic-type macroscopic equations for microscopic models and with the study of the nature of nonequilibrium ordering and phase transitions in condensed matter physics. The present study concerns the latter problem.

The resulting phase diagram for the stable (nonequilibrium) homogeneous steady states of that generalized model is very rich and interesting, even for a given choice of the rates for the reaction and diffusion processes (the choice by Metropolis *et al.*⁽¹⁶⁾). As a matter of fact, while the limit $p \rightarrow 1$ and $T' \rightarrow \infty$ represents pure mean-field behavior and lack of correlations, the system undergoes a variety of first- and second-order phase transitions as one samples the parameters p , T , and T' . That is, for small values of the velocity p of the disturbing diffusion process, say for $p < 0.7$, the system always displays (for appropriate pairs of values of the two relevant temperatures) a continuous phase transition with Onsager critical exponents, and other quantities may straightforwardly be scaled to the equilibrium ones. When p is large enough, however, the system may undergo as well discontinuous phase transitions of several kinds. Namely, for a fixed large value of p , the transition at low reaction temperatures T is first order with strong discontinuities when T' remains small (say $T' \approx 0.1T^0$, where T^0 is the Onsager critical temperature), while for large T' (say $T' \approx 10T^0$) it is only weakly first order in some quantities. Furthermore, while the segregated states for small T' resemble those encountered in the simulation of spinodal phenomena, which are positively characterized by long-range order, those for large T' rather parallel the ones in nucleation phenomena, where the order is relatively shorter. Also, the phase transition at low T is second order when $T' \approx T^0$, and the critical exponents are observed to change over those of the Landau classical theory as one tends to increase T' trying to reach the region of the phase diagram where the phase transition becomes discontinuous.

The present model system clearly deserves more attention, once the

limits $\Gamma \rightarrow \infty$ and/or $T' \rightarrow \infty$ are relatively well understood and one has evidence of a rich structure under more general conditions. We are presently carrying out further MC analysis, and also the study of the same model with a kinetic cluster-variation technique; we expect in particular to obtain some insight about the influence of the rates $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$ and $c(\mathbf{s}; \mathbf{x})$ on the phase diagram.

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