

Reaction-diffusion lattice gas: Theory and computer results

J. J. Alonso and J. Marro

Departamento de Física Aplicada, Facultad de Ciencias, Universidad de Granada, E-18071 Granada, Spain

J. M. González-Miranda

Departamento de Física Fundamental, Facultad de Física, Universidad de Barcelona, E-08028 Barcelona, Spain

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We report on the study of nonequilibrium ordering in the reaction-diffusion lattice gas. It is a kinetic model that relaxes towards steady states under the simultaneous competition of a thermally activated creation-annihilation (*reaction*) process at temperature T , and a diffusion process driven by a heat bath at temperature $T' \neq T$. The phase diagram as one varies T and T' , the system dimension d , the relative *a priori* probabilities for the two processes, and their dynamical rates is investigated. We compare mean-field theory, new Monte Carlo data, and known exact results for some limiting cases. In particular, no evidence of Landau critical behavior is found numerically when $d=2$ for Metropolis rates but Onsager critical points and a variety of first-order phase transitions.

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I. MODEL

We report on the study of ordering in a nonequilibrium lattice gas whose configuration, $\mathbf{s} = \{s_{\mathbf{x}} = \pm 1; \mathbf{x} \in \mathbb{Z}^d\}$, where the lattice dimension is either $d = 1$ or 2 , evolves in time owing to the combination of two processes. Namely, \mathbf{s} changes stochastically owing to both creation-annihilation (referred to as *reaction*) processes activated by a heat bath at temperature T , and *diffusion* processes consisting of nearest-neighbor (NN) exchanges driven by a heat bath at temperature T' instead. The two processes are independent in continuous time, with p the *a priori* probability of exchanges per bond and $(1-p)$ that of reactions per site. Let us denote by \mathbf{L}_G the *Glauber operator* [1] that produces stochastic changes of $s_{\mathbf{x}}$ to $-s_{\mathbf{x}}$ with probability per unit time (*rate*) $c(\mathbf{s}; \mathbf{x})$, and by \mathbf{L}_K the *Kawasaki operator* [2] that induces stochastic interchanges of the occupation variables at NN sites \mathbf{x} and \mathbf{y} with rate $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$. The probability of \mathbf{s} at time t , $\mu(\mathbf{s}; t)$, is governed by the (Markovian) master equation

$$\frac{d\mu(\mathbf{s}; t)}{dt} = [p\mathbf{L}_K + (1-p)\mathbf{L}_G]\mu(\mathbf{s}; t). \tag{1.1}$$

The transition rates $c(\mathbf{s}; \mathbf{x})$ and $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$ satisfy detailed balance with respect to different temperatures. This has two main, well-known consequences. On the one hand, $\mathbf{L}_K\mu_{\text{eq}}^{T'}(\mathbf{s}; m) = 0$, with

$$\mu_{\text{eq}}^{T'}(\mathbf{s}; m) = Z(\hat{\mu})^{-1} \exp \left[-H(\mathbf{s})/k_B T' + \hat{\mu} \sum_{\mathbf{x}} s_{\mathbf{x}} \right], \tag{1.2}$$

where $\hat{\mu} = \hat{\mu}(m)$, $m = N^{-1}(d/d\hat{\mu}) \ln Z(\hat{\mu})$, and $Z(\hat{\mu}) \equiv \sum_{\mathbf{s}} \exp[-H(\mathbf{s})/k_B T' + \hat{\mu} \sum_{\mathbf{x}} s_{\mathbf{x}}]$. Here $H(\mathbf{s})$ represents the (configurational) energy, which we assume hereafter to be

$$H(\mathbf{s}) = -J \sum_{\text{NN}} s_{\mathbf{x}} s_{\mathbf{y}} \tag{1.3}$$

for simplicity. On the other hand, $\mathbf{L}_G\mu_{\text{eq}}^T(\mathbf{s}) = 0$, with

$$\mu_{\text{eq}}^T(\mathbf{s}) = \left[\sum_{\mathbf{s}} \exp[-H(\mathbf{s})/k_B T] \right]^{-1} \exp[-H(\mathbf{s})/k_B T]. \tag{1.4}$$

Consequently, (1.1) has two limiting cases that are a canonical reference for the (nonequilibrium) situations of interest here. Namely, the system reduces to the Glauber or kinetic Ising model (which has a nonconserved order parameter) [1] for either $p \equiv 0$ or $c(\mathbf{s}; \mathbf{x}, \mathbf{y}) \equiv 0$. Then, independently of $c(\mathbf{s}; \mathbf{x})$, the asymptotic solution of (1.1) is the Gibbs state (1.4) for temperature T and energy $H(\mathbf{s})$. It is known to undergo (in the infinite-volume limit that is considered throughout the paper, except for Monte Carlo computations) a second-order phase transition at temperature $T_C^0 \geq 0$, where equality holds for $d = 1$. On the other hand, one recovers the (kinetic) Kawasaki lattice-gas model (which corresponds to the Ising model with a conserved order parameter) [2] for either $p \equiv 1$ or $c(\mathbf{s}; \mathbf{x}) \equiv 0$. Independently of $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$, (1.1) leads to the Gibbs state (1.2) at temperature T' with a fixed value m for the order parameter. A critical point exists at $T_C'^0 \geq 0$ when $m = 0$, and first-order phase transitions arise when $m \neq 0$.

The situation is very different otherwise: The steady state $\mu_{\text{st}}(\mathbf{s})$, defined as the limit of $\mu(\mathbf{s}; t)$ in (1.1) as $t \rightarrow \infty$, is away from equilibrium in general. This is similar to having \mathbf{s} acted on by some external non-Hamiltonian agent. In fact, $\mu_{\text{st}}(\mathbf{s})$ cannot be described, in general, by a Hamiltonian with suitable properties, e.g., one that is short ranged and plays in the expression for $\mu_{\text{st}}(\mathbf{s})$ the role played by $H(\mathbf{s})$ in (1.2) or (1.4) [3]. This implies, in particular, that $\mu_{\text{st}}(\mathbf{s})$ may depend, even qualitatively, on the specific form assumed for the reaction and diffusion rates. Under those nonequilibrium conditions, the system may exhibit ordering phenomena. The latter may be denoted as *nonequilibrium phase transitions*, given that

some (nonrigorous) similarity exists with an equilibrium situation in which thermodynamic instabilities occur. It is reasonable to expect a dependence of steady states and phase transitions on the values of the model parameters, namely, $d, p, J, T, T', c(\mathbf{s}; \mathbf{x})$, and $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$. Thus, nonequilibrium phase transitions (and associated critical behavior) are expected to be more varied than for equilibrium states, given the singular simplicity of the latter.

The preceding discussion suggests a theoretical interest to investigate the implications of (1.1)–(1.4) as a well-defined model in nonequilibrium statistical mechanics, where a general formalism such as the powerful Gibbs ensemble theory is lacking. In addition, reaction-diffusion systems have a practical interest in physics, chemistry, and biology, for example [4]. Consequently, some limiting cases of the generalized model defined above have been analyzed in detail in recent years (Refs. [4–15], and references therein). Further interest ensues from the fact that it has been shown rigorously to have a macroscopic, hydrodynamiclike description within the limit $T' = \infty$ and $p \rightarrow 1$ [5–7]. For instance, the order parameter $m(\mathbf{r}, t) \equiv \langle s_{\mathbf{x}} \rangle_m$ satisfies the *reaction-diffusion equation* $\partial m(\mathbf{r}, t) / \partial t = \frac{1}{2} \nabla^2 m(\mathbf{r}, t) + F\{m(\mathbf{r}, t)\}$ for the most random and microscopically fastest diffusion process (if time and space are appropriately rescaled in the model); here $\mathbf{r} \in \mathbb{R}^d$, $F\{m\} = -2 \langle s_{\mathbf{x}} c(\mathbf{s}; \mathbf{x}) \rangle_m$, and the averages are taken with respect to a Bernoulli state with uniform m . This equation may then be used to study [5,9] (nonequilibrium) homogeneous steady states, i.e., solutions of $F\{m\} = 0$, for example. It has confirmed, in particular, the existence of phase transitions with no equilibrium counterpart, e.g., in a one-dimensional system for some realizations of $c(\mathbf{s}, \mathbf{x})$.

For completeness, Sec. VI contains a description of phase transitions and critical phenomena in the reaction-diffusion lattice gas as obtained from both previous exact, mean-field and Monte Carlo (MC) studies [4–15], and our findings in the present paper. The case with $0 < p < 1$ and/or finite T' has been studied so far only partially by mean-field [8] and/or MC methods [10,14]. Consequently, a detailed investigation for general choices of the relevant parameters, including the consideration of further reaction and diffusion rates, is desirable. As a matter of fact, the case of finite T' is the most general and interesting one for practical purposes. We discuss in this paper the main results from a systematic study in that direction. Namely, we report in Sec. II MC data for $d = 2$ that allow definite conclusions concerning phase diagrams and critical behavior. Section III describes the main equations and results from a kinetic mean-field theory. It is applied in Sec. IV to study steady states for $d = 1$, $T, T' \in [0, \infty]$, varying values of p , and rather arbitrary functions for $c(\mathbf{s}; \mathbf{x})$ and $c(\mathbf{s}; \mathbf{x}, \mathbf{y})$; moreover, our equations are for both signs of J in (1.3). Section V is devoted to the mean-field study of a square lattice. A comparison between analytical and computer-simulation results is made throughout the paper.

II. COMPUTER SIMULATION RESULTS FOR $d = 2$

We analyze in this section a series of computer simulations that notably extend the data reported before for

$d = 2$ [14]. The basic Monte Carlo step is as follows. After a site \mathbf{x} is chosen at random, either $s_{\mathbf{x}}$ is exchanged (with probability p) with one of its NN $s_{\mathbf{y}}$ using the rate $c(\mathbf{s}; \mathbf{x}, \mathbf{y}) = \min[1, \exp(-\delta H / k_B T')]$, or $s_{\mathbf{x}}$ is changed to $-s_{\mathbf{x}}$ (with probability $1-p$) according to the rate $c(\mathbf{s}; \mathbf{x}) = \min\{1, \exp(-\delta H / k_B T)\}$. Here δH is the change of (1.3) associated to the attempted transition. The motivation for assuming in this section that both diffusion and reaction processes are governed by the Metropolis algorithm [16] is that it turns out in practice most efficient when trying to simulate stabilized condensation disturbed by diffusion. The choice of rates influences, in general, the steady state, but it is more easily evaluated in our analytical treatments below.

We have considered square lattices with $L^2 \equiv N$ sites, periodic boundary conditions, and attractive ($J > 0$) NN interactions. Most results are for either $L = 64$ or 128 ; relatively long evolutions, typically between 5×10^4 and 10^6 MC steps (per lattice site), were performed to assure confident statistics. This amounts to a notable addition to the data reported in Ref. [14], e.g., a finite-size scaling analysis with L ranging from 8 to 128 is employed in the investigation of critical behavior here.

In steady conditions, we have monitored the order parameter or *magnetization*,

$$m = N^{-1} \left\langle \sum_{\mathbf{x}} s_{\mathbf{x}} \right\rangle, \quad (2.1)$$

where $\langle \rangle$ represents the MC average over configurations, the energy,

$$e = -(2JN)^{-1} \langle H \rangle, \quad (2.2)$$

the squared mean fluctuations of m and e ,

$$C = N^{-1} (\langle H^2 \rangle - \langle H \rangle^2), \quad (2.3)$$

$$X = (J/k_B) \left[\left\langle \left| \sum_{\mathbf{x}} s_{\mathbf{x}} \right|^2 \right\rangle - \left\langle \left(\sum_{\mathbf{x}} s_{\mathbf{x}} \right)^2 \right\rangle \right], \quad (2.4)$$

and the short-ranged order parameter [17]

$$\sigma = \langle N_{+-} + N_{--} (N_{+-})^{-2} \rangle, \quad (2.5)$$

where N_{+-} , N_{++} , and N_{--} are the number of the three different NN pairs, i.e., particle-hole, particle-particle, and hole-hole, that may occur in the system, respectively. We have checked that σ is in practice indistinguishable from $\langle N_{++} N_{--} \rangle \langle N_{+-} \rangle^{-2}$, as one may expect. It may be shown that the critical behavior of σ is then characterized by the familiar exponents α and β ; namely, one gets

$$\sigma = \sigma_c + A \epsilon^{1-\alpha} - B \epsilon^{2\beta}, \quad (2.6)$$

where A and B are both positive constants, $\epsilon \equiv 1 - T/T_C$, T_C is the critical temperature, and σ_c is nonsingular. The study of σ is convenient because it behaves more simply and smoothly than more standard quantities. In particular, σ has a simple scaling behavior with N and, according to (2.6) and further evidence [17,18], it is expected to exhibit in general either a well-defined peak at T_C or else a monotonic variation around T_C ; the latter corresponds to the classical values $\beta = \frac{1}{2}$ and $\alpha = 0$.

The preliminary data in Ref. [14] already suggested that the MC analysis becomes easier when one recognizes the existence of three distinct types of qualitative behavior: Type I looks similar to an equilibrium second-order phase transition. It occurs when the reaction temperature has a value T_C that is a function of both p and T' ; cf. Table I. This is illustrated by the circles in Figs. 1, 2(a), and 3, which reveal the following general behavior. The energy [Fig. 1(a)] and the magnetization [Fig. 1(b)] have a continuous variation with T ; σ (Fig. 2) tends to describe a

peak that becomes more acute as N is increased; the fluctuations C [Fig. 1(c)] and X [Fig. 1(d)] suggest divergences at both sides of T_C ; and σ and X are highly asymmetrical around T_C , in contrast with the apparent symmetry of C . This seems to be the most general behavior, as indicated in Table I and Fig. 3. It is observed for not too large values of p , say for $p \leq 0.8$, for $p = 0.85$ as far as $\tau' \equiv T'/T_C^0 < 7$ (where $T_C^0 = 2.27J/k_B$ is the Onsager critical temperature), and for $p = 0.95$ excluding the ranges $\tau' < 1$ and $\tau' > 5.2$. Consistent with that trend,

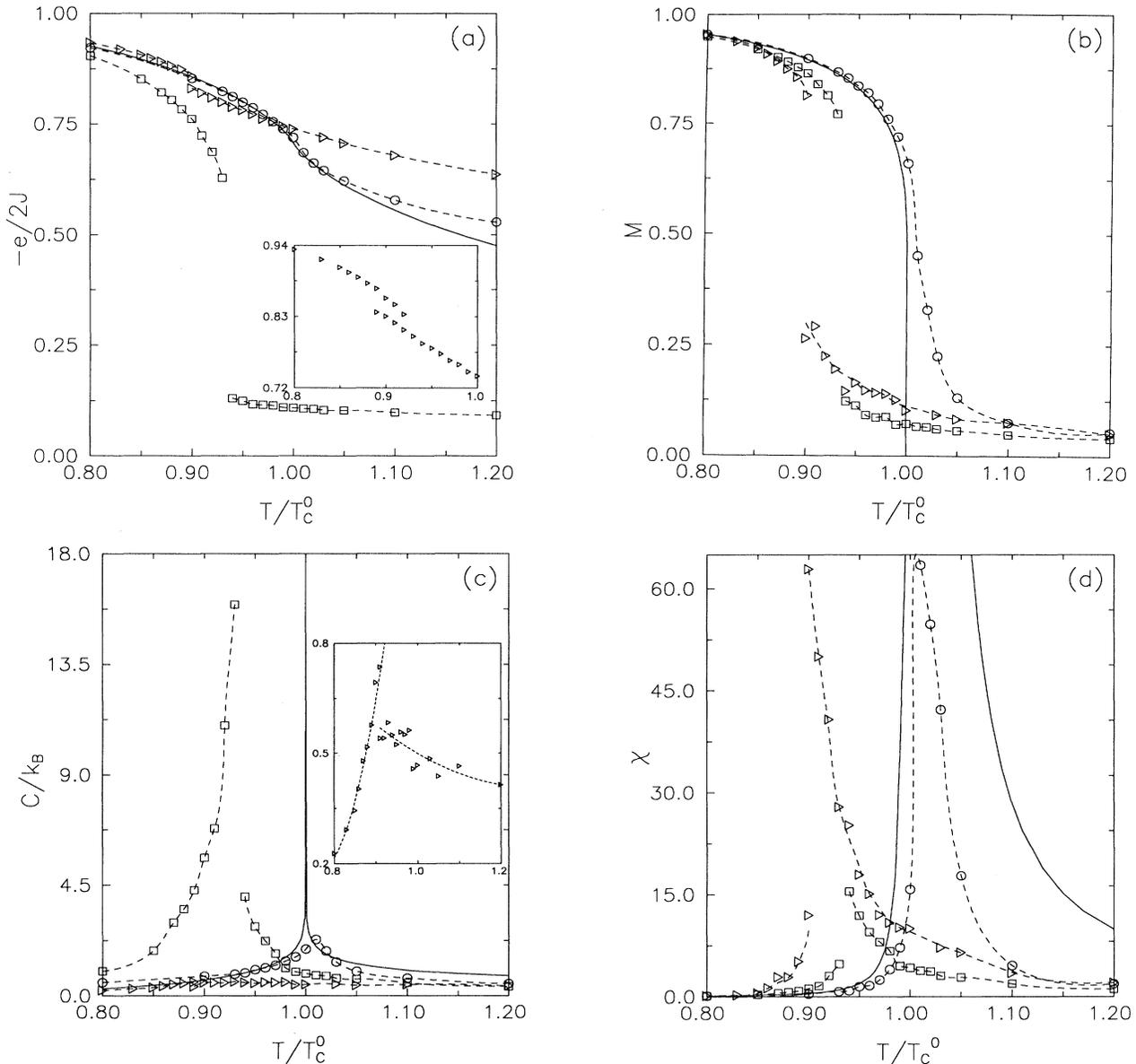


FIG. 1. (a) The energy defined in (2.2), as a function of the reaction temperature (in units of the Onsager temperature, $T_C^0 = 2.27J/k_B$), for $p = 0.95$ and different values of the diffusion temperature, $\tau' \equiv T'/T_C^0 = 0.1$ (triangles), 1 (circles), and 10 (squares), as obtained by the MC method for the two-dimensional system. The dashed lines are guides to the eye. The solid line corresponds to the equilibrium (infinite-volume) case. The inset amplifies the case $\tau' = 0.1$ around the transition temperature; this reveals some metastable states. (b) The same but for the magnetization order parameter as defined in (2.1). (c) The same but for the energy fluctuations as defined in (2.3). (d) The same but for the magnetization fluctuations X as defined in (2.4).

type I was reported for $p < 0.83$ when $\tau' = \infty$ [9]. The facts that the data for e and m can be mapped onto a single curve by simply representing them versus $T/T_c(p, T')$, and that such a common curve agrees (except for finite-size effects) with the Onsager solution, are noteworthy. Moreover, as depicted by Fig. 3(a) for example, the cases $\tau' > 1$ and $\tau' < 1$ are not equivalent; in fact, there is no reason why $\tau' = 1$ should have any special significance. The general agreement between the nonequilibrium C and X and the corresponding curves at equilibrium tends to get worse the further one departs from $\tau' = 1$, however; it is evident for both $\tau' \gg 1$ and $\tau' \ll 1$.

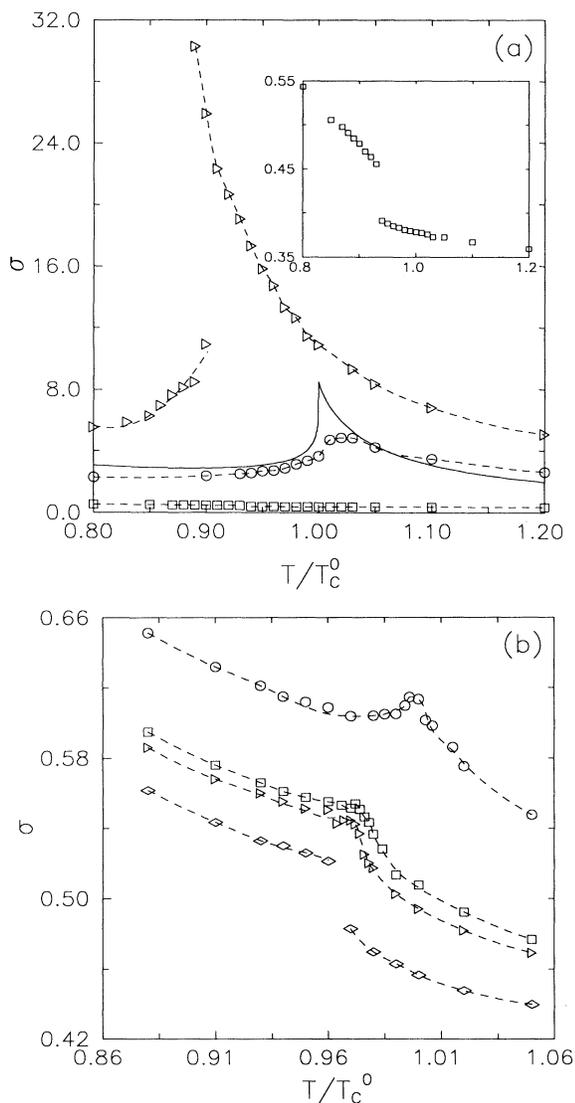


FIG. 2. Short-ranged order parameter (2.5). (a) The same cases and symbols as in Fig. 1(a); the inset amplifies the case $\tau' = 10$. The solid line represents the exact equilibrium result corresponding to $\sigma = \frac{1}{4}[(1+e)^2 - 4m^2](1-e)^{-2}$ [17]. (b) Data for different phase points at $p = 0.95$ as one approaches the tricritical point ($\tau' \approx 5.35$); namely, for $\tau' = 4, 5, 5.2$, and 6 from top to bottom.

Such a departure from a common behavior in some occasions seems related to the breakdown of the fluctuation-dissipation relations.

Summing up, *type I* resembles in many respects the (equilibrium) case for the Glauber and Kawasaki models, but nonequilibrium features are enhanced as τ' is increased; large values of p , say $p > 0.85$, and small values of τ' , say $\tau' < 1$, also tend to induce strong, qualitative departures from equilibrium behavior; cf. below.

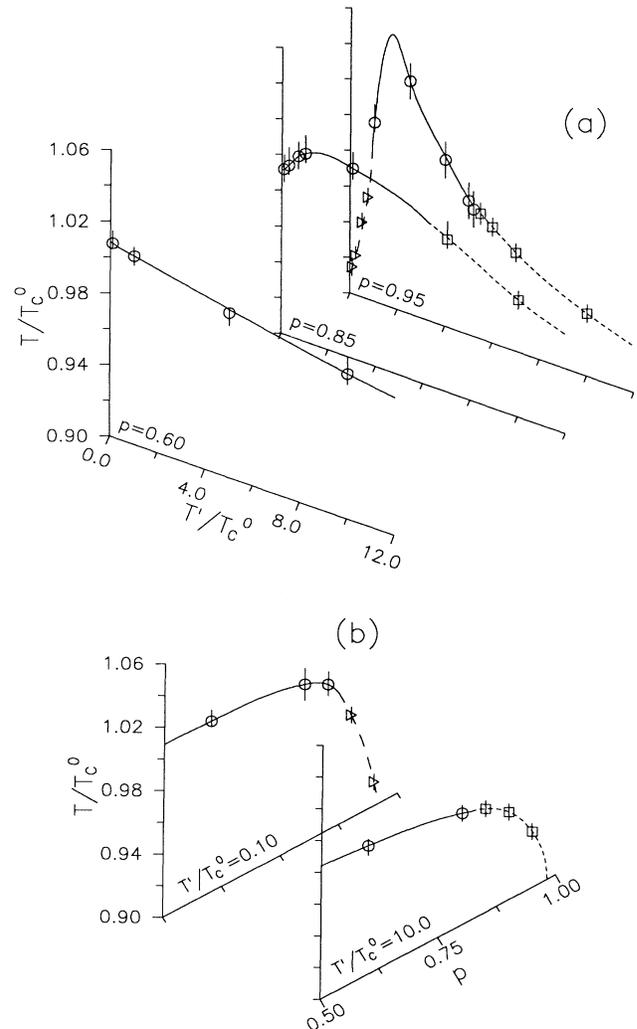


FIG. 3. Several sections, as indicated, of the phase diagram that follows from the MC analysis of the two-dimensional system, with both reaction and diffusion rates implemented by the Metropolis algorithm, assuming temperatures T and T' , respectively. The nature of the phase transition occurring at the sampled phase points is represented by using circles, squares, and triangles, respectively, for *types I, II, and I'* (cf. Sec. II). Error bars are indicated. The solid lines represent lines of critical points; the dashed lines correspond to discontinuous phase transitions; both are obtained as cubic (spline) fits to the data. Note that a perspective is simulated in the graphs, trying to suggest the phase diagram surface.

TABLE I. MC estimations for transition temperatures as a function of T' and p for $d=2$, corresponding to the graphs in Fig. 3, and type of behavior at each phase point, as described in the main text. Temperatures are in units of the Onsager critical temperature. Error bars are always smaller than ± 0.01 . The case $T' = \infty$ corresponds to the exact results in Ref. [9].

T'	p					
	0.60	0.80	0.85	0.90	0.95	1
0.10	1.008,I	1.000,I	0.992,I	0.965,II'	0.915,II'	
0.25					0.922,II'	
0.30			0.995,I			
0.50					0.942,II'	
0.70			1.002,I			
0.75					0.957,II'	
1	1.005,I		1.005,I		1.000,I	
2.5					1.030,I	
3			1.006,I			
4					0.993,I	
5	0.992,I				0.975,I	
5.2					0.971,I	
5.5					0.970,II	
6					0.965,II	
7			0.985,II		0.955,II	
10	0.981,I	0.970,I	0.965,II	0.955,II	0.935,II	
∞	0.96,I	0.94,I	0.92,II	II	0.90,II	0.855,II

Type II is characterized by well-defined discontinuities in e , m , C , and X at some reaction temperature that is a function of p and T' (cf. Table I). This is illustrated by the squares in Figs. 1, 2(a), and 3. The parameter σ has no peak, but monotonically decreases with increasing reaction temperature, and shows a clear discontinuity at the transition temperature. Figure 2(a) reveals also that σ is notably smaller than for *type I*. This kind of behavior, which is the rule for $p \geq 0.85$ when $\tau' > 7$, resembles an equilibrium first-order phase transition. It is reinforced by the fact that the system evolves near the transition via metastable states that typically decay to the steady state in a time of order 10^4 to 10^5 MC steps. The existence of metastable states is illustrated in Fig. 4. The direct inspection of configurations suggests the presence of coexisting phases; no systematic MC analysis of this was performed, but it is suggested also by mean-field theory; cf. Sec. V. *Type II* behavior has also been reported in Ref. [9] when $\tau' = \infty$ for $p > 0.83$.

The situation also resembles a discontinuous phase transition for $p = 0.95$ and $\tau' < 1$, but it has some qualitative differences with *type II*. In particular, the MC analysis is hampered now by the systematic presence of very slow evolutions, in addition to long-lived metastable states that last 5×10^5 MC steps or more. We shall denote this as *type II'* behavior. It is identified by triangles in Figs. 1–3, which reveal the following. As for *type II*, a discontinuity in m exists [cf. Fig. 1(b)], while the discontinuities in e [Fig. 1(a)] and C [Fig. 1(c)] are much weaker, and the structure of X [Fig. 1(d)] near the transition temperature (which depends on p and T' ; cf. Table I) is much more pronounced. The behavior of σ in Fig 2(a)

is also remarkable. It suggests a discontinuity, but its shape more resembles the equilibrium case than *type II*. Moreover, σ is much larger than for *types I* and *II*; i.e., a larger degree of local order is also evident by direct inspection of particle configurations [14]. The fact that the evolutions for *type II'* are extremely slow produces in practice a more noisy data than usual for C and X [cf. inset for Fig. 1(c)].

The rich behavior of the system is illustrated by the case $p = 0.95$ in Fig. 3 and Table I. In fact, although the transition temperature (which exhibits a well-defined maximum at $\tau' \approx 2.5$) never differs by more than 10% from the Onsager value, the phase transition changes there from *type II'* to *type I*, and then to *type II*, as τ' is increased from zero. The changes between types are smooth in our finite system (as suggested, for example, by the values in Table I), but one should probably expect the existence of two tricritical points for the infinite-volume system. If they are denoted by $T_1(p)$ and $T_2(p)$, respectively, the data indicate $T_1(0.95) \approx 0.8$, $T_1(0.9) > 0.1$,

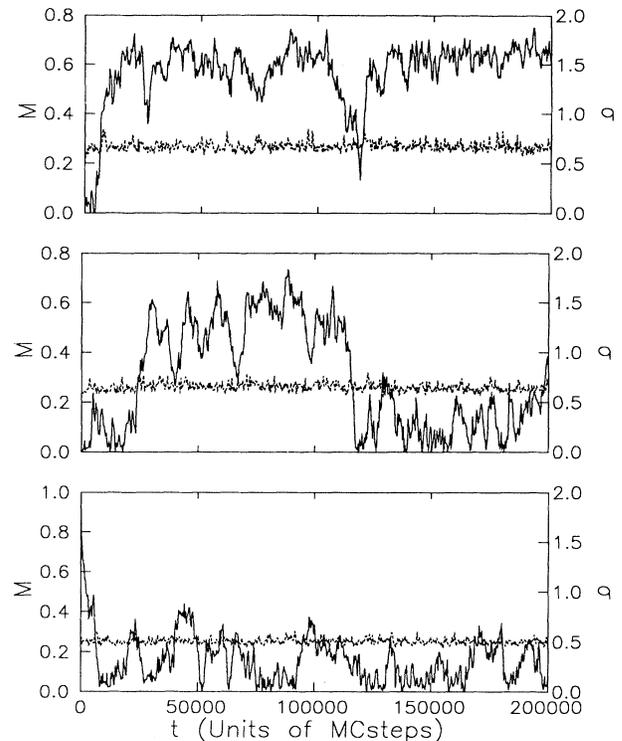


FIG. 4. Typical MC time evolutions for the magnetization (the curve with larger fluctuations) and short-range order parameter (the curve in which fluctuations have a relatively smaller amplitude) for a *type II* phase transition near the transition point. The data correspond to $p = 0.95$ and reaction temperatures $T/T_c^0 = 0.964, 0.968$, and 0.973 , respectively, from top to bottom. Thus, the top case is inside the error bars for the corresponding transition temperature, and the temperature differences are around 0.5% between two consecutive cases. Note the metastability revealed by the second case. The energy behaves similar to m here.

$T_1(p \leq 0.85) \approx 0$, $T_2(0.95) = 5.35 \pm 0.05$, $T_2(0.85) < 7$, and $T_2(p < 0.85) \approx \infty$. The simplest picture emerges for $p = 0.6$, where only *type I* has been detected for $0 < \tau' \leq \infty$. The critical temperature in this case decreases a little bit faster than linearly with increasing τ' . The fact that the transition is always of second order for small p , while it becomes of first order (the type of discontinuities changing with τ') for larger p , is consistent with the reported existence of a tricritical point at $p = p_t \approx 0.83$ when $\tau' = \infty$ [9,10].

The critical behavior is a specific question. The following has been stated concerning the various second-order phase transitions in the system: (1) when $p < p_t$ and the diffusion temperature is infinite, they seem characterized by Onsager critical exponents [10], as for $p = 0$, in agreement with some theoretical arguments [19]; (2) the situation is less clear-cut as $p \rightarrow p_t$ [14]; and (3) the critical behavior for *type I* when τ' is finite is *roughly consistent* with the Onsager case [14]. We have investigated systematically the critical indexes, including $m^{1/\beta}$ -versus- T plots (Fig. 5) and standard [20] finite-size scaling analysis (Figs. 6 and 7), trying to reach more definite conclusion. The main conclusion is that critical behavior is indistinguishable indeed from the equilibrium one. It is true even near tricritical points separating *type I* from *type II*, i.e., as $p \rightarrow p_t$. This is illustrated by Figs. 5–7, and it also follows from the detailed study of σ [cf. Fig. 2(b)]. The much more limited data reported earlier [14] apparently suggested a changeover of β from the Onsager value $\frac{1}{8}$ to the Landau classical one $\frac{1}{2}$, in agreement with some expectations. It was based on the observation that σ apparently had a tendency towards a monotonic variation as $p \rightarrow p_t$. In particular, a gradual smoothing of the peak that, as indicated above, corresponds to nonclassical critical behavior was observed. We may conclude with

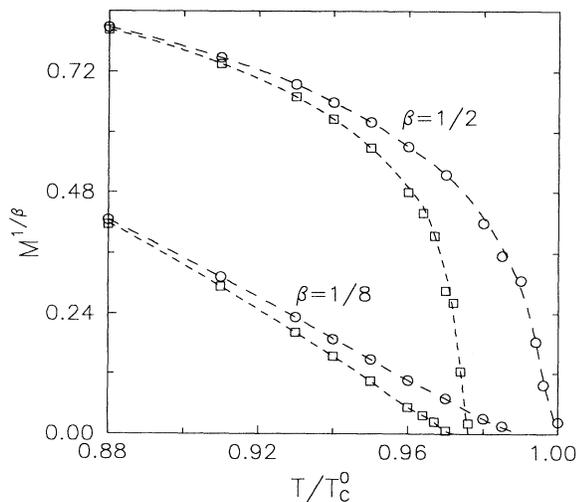


FIG. 5. Plot of $m^{1/\beta}$ versus T/T_c^0 for $\beta = \frac{1}{2}$ and $\beta = \frac{1}{8}$ (as indicated). The data correspond to an extrapolation to the infinite lattice with $p = 0.95$ and $\tau' = 4$ (circles) and 5.2 (squares); the latter is within 3% of the tricritical point ($\tau' \approx 5.35$).

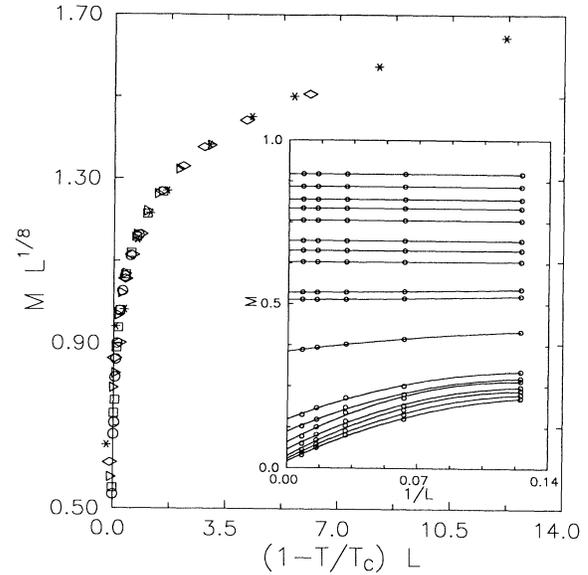


FIG. 6. Standard finite-size scaling plots (assuming $\beta = \frac{1}{8}$ and $\nu = 1$) of the magnetization data for $p = 0.95$ and $\tau' = 5.2$. The symbols in the main graph correspond to $L = 8$ (circles), 16 (squares), 32 (triangles), 64 (rhomboids), and 128 (asterisks).

confidence, however, that it was only an artifact related to finite-size effects and, more decisively, to the relatively small magnitude of σ near p_t ; cf. Fig. 2(b).

Finally, we have monitored the spatial relaxation of site-site correlation functions at high temperature. They decay exponentially at large distances, as expected for a nonconserved magnetization [21], in agreement also with some analytical computations [10]. This behavior is illustrated by Fig. 8 and, in particular, by the inset therein.

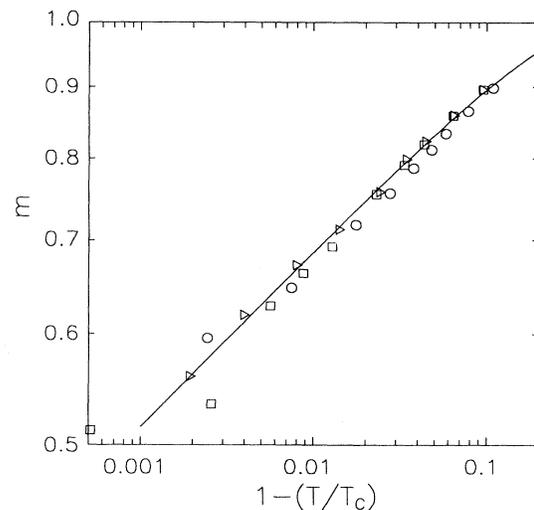


FIG. 7. Log-log plot of the magnetization versus $1 - T/T_c(T, p)$ for $p = 0.95$ and $\tau' = 4$ (circles), 5 (triangles), and 5.2 (squares). The solid line corresponds to the Onsager solution.

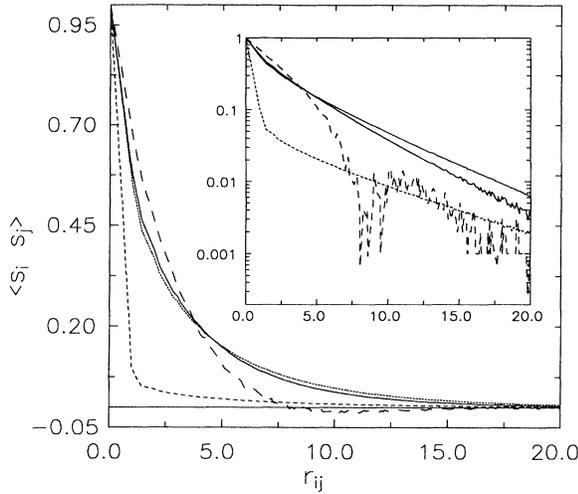


FIG. 8. Site-site correlation functions for $p=0.95$ and $r'=0.1$ (long dashes), 1 (solid line), and 10 (short dashes), and for the Ising model (dotted line), when the reaction temperature is $T=1.10T_C^0 > T_C(T', p)$. The inset is a semilogarithmic plot of the absolute values of the same data.

III. THE ANALYTICAL METHOD, AND SOME GENERAL RESULTS

Our main approach in Secs. 3–5 is a field theory for kinetic lattice systems [22]. Namely, (1.1) is reduced to a few equations for the time evolution of mean local quantities by neglecting long-wavelength fluctuations and restricting correlations to those between a small number of sites. A small, compact domain of sites, say D , which allows for the involved dynamical process is considered. The correlations between the particles in D and those in the rest of the system are suppressed. With this aim, one neglects fluctuations at the surface of D in such a way that the resulting description is a first-order mean-field one. The method generalizes the (equilibrium) theories of Bethe and Peierls to a class of kinetic phenomena. The minimum size of D that allows for the competition between reaction and diffusion consists of two NN sites (the *interior* of D , denoted I) surrounded by its respective NN sites outside I (the *surface* of D , denoted S); the number of sites that belong to S is $2(2d-1)$. On the other hand, the only correlations that are allowed to first order are pair (NN) ones between I and S . Consequently, the (approximate) description of the configuration at any time only requires the consideration of the particle density, denoted $n \equiv N_+/N$, and of the density of (NN) particle-particle pairs, $z \equiv N_{++}/dN$. It is helpful, however, to refer also to $v \equiv N_-/N = 1-n$, $w \equiv N_{--}/dN = 1+z-2n$, and $2v \equiv N_{+-}/dN = 2(n-z)$. The magnetization (2.1) is $m = n - v$, and the energy (2.2) is $e = 1 - 4v$. A higher-order approximation would involve both a larger size for D , further correlations, and, consequently, more variables.

Our approach is similar to the one used before to study the limiting case $T' = \infty$ with Metropolis rates [8]. It is convenient in the present situation, given that mean-field

approximations are typically believed to be more relevant for nonequilibrium than for equilibrium phenomena. In fact, the case $p \rightarrow 1$ (with $T' = \infty$) has a mean-field nature, as mentioned above, and it is concluded here that classical behavior occurs in other circumstances as well. The method is also convenient because it involves an explicit description of time evolution, steady states and stability conditions, and one may consider different values for any of the relevant model parameters. In particular, one may apply the method to any choice of transition rates. It is interesting to consider a generalized rate for transitions from s to s' , given as

$$c(s'|s) = f(\delta H), \quad \delta H \equiv [H(s') - H(s)](k_B \Omega)^{-1}, \quad (3.1)$$

where $\Omega = T$ or T' for reaction or diffusion, respectively, and $f(X)$ is arbitrary, except that the detailed balance condition,

$$c(s'|s)/c(s|s') = \exp(-\delta H), \quad (3.2)$$

is assumed to hold for simplicity. We refer below to the rates introduced to deal with different problems by Metropolis *et al.* [16], $f(X) = \min\{1, e^{-X}\}$; by Kawasaki [2], $f(X) = [1 + e^X]^{-1}$; by van Beijeren and Schulmann [23], $f(X) = e^{-(1/2)X}$; and by De Masi, Ferrari, and Lebowitz [5], $f(X) = e^{-(1/2)X} [\cosh(J/k_B \Omega)]^{-2d}$. The specific notation $c(s; \mathbf{x}) = \phi(X)$ and $c(s; \mathbf{x}, \mathbf{y}) = \psi(X)$ for $f(X)$ is used hereafter to differentiate the two elementary processes.

The configurations of the *minimum cluster* D (as defined above) are characterized by the occupation variables at the two interior sites (to be denoted s_j , $j=1,2$), and by the number i_j ($\leq 2d-1$) of (surface) sites that are nearest neighbors of j and have occupation variables that equal +1. The associated probabilities are

$$Q_D(s_1, s_2; i_1, i_2) = p(s_1, s_2) \prod_{j=1}^2 \binom{2d-1}{i_j} p(+|s_j)^{i_j} p(-|s_j)^{2d-1-i_j}. \quad (3.3)$$

Here $p(s|s') = p(s, s')/p(s')$ is a conditional probability, $p(s)$ is the probability that the occupation variable at a given site equals s , so that $p(+)=1-p(-)=n$, and $p(s, s')$ is the probability that a given NN pair has occupation variables (s, s') , i.e., $p(+, +)=z$, $p(-, -)=w$, and $p(+, -)=p(-, +)=v$. The equations governing n and z to first order follow when one recognizes that dynamics consists of a competition of: (A) interchanges at temperature T' with probability $p\psi(\delta H)$, and (B) reactions at temperature T with probability $(1-p)\phi(\delta H)$. Let us consider separately the action of A and B. By the philosophy of the master equation, the variation with time of α ($\equiv n$ or z) that is induced by the action of (only) mechanism B is

$$\frac{d\alpha}{dt} = \sum_{s_1, s_2 = \pm 1} \sum_{i_1, i_2 = 0}^{2d-1} \delta\alpha Q_D(s_1, s_2, i_1, i_2) \phi(\delta H), \quad (3.4)$$

where $\delta\alpha$ represents the change of α caused by the inver-

sion of s_1 ; the fact that the method involves spatial homogeneity, which implies the same action on s_2 as on s_1 , has been used. The sum over i_2 in (3.4) only affects the probability (3.3); thus,

$$\frac{d\alpha}{dt} = \sum_{s_1=\pm 1} \sum_{i=0}^{2d} \delta\alpha Q_D(s_1, i) \phi(\delta H), \quad (3.5)$$

where

$$Q_D(s_1, i) \equiv \binom{2d}{i} p(s_1) p(+|s_1)^i p(-|s_1)^{2d-i}. \quad (3.6)$$

Equations (3.5) and (3.6) are precisely what one would have obtained by considering mechanism B and the corresponding minimum cluster, i.e., only one interior site (and its NN surface sites). It follows from (3.5) that

$$\frac{dn}{dt} = F(n, z; T) \equiv \sum_{i=0}^{2d} \binom{2d}{i} v^i \left[\frac{w^{2d-i}}{v^{2d-1}} - \frac{z^{2d-i}}{n^{2d-1}} \right] \times \phi(d-i), \quad (3.7)$$

where $\phi[s_1(i-d)] \equiv \phi[4J s_1(i-d)/k_B T]$, for $\alpha \equiv n$ ($\delta n = -s_1$), and

$$(4d-1) \frac{dz}{dt} = G(n, z; T) \equiv \sum_{i=0}^{2d} \binom{2d}{i} v^i \left[\frac{i w^{2d-i}}{v^{2d-1}} - \frac{(2d-i) z^{2d-i}}{n^{2d-1}} \right] \times \phi(d-i), \quad (3.8)$$

for $\alpha \equiv z$ [$\delta z = -i s_1 (4d-1)^{-1}$]. Next, consider the action of mechanism A on the original *minimum cluster* D, i.e., the interchange $s_1 \rightleftharpoons s_2$ at the interior sites. This leaves r constant, but changes z and H by $s_1(i_2 - i_1)(4d-1)^{-1}$ and $4J s_1(i_1 - i_2)/k_B T'$, respectively, for $s_1 = -s_2$. Consequently, one gets under the same philosophy that

$$(4d-1) \frac{dz}{dt} = K(n, z; T') \equiv 2 \sum_{i_1, i_2=0}^{2d-1} \binom{2d-1}{i_1} \binom{2d-1}{i_2} (i_2 - i_1) \times \frac{z^{i_1} w^{2d-1-i_2} v^{2d-i_1+i_2}}{(n v)^{2d-1}} \psi(i_1 - i_2), \quad (3.9)$$

where a sum over s_1 has been performed. Finally, one obtains

$$\frac{dn}{dt} = (1-p)F(n, z; T), \quad (3.10)$$

$$(4d-1) \left[\frac{dz}{dt} \right] = (1-p)G(n, z; T) + pK(n, z; T'), \quad (3.11)$$

where $F(T)$, $G(T)$, and $K(T')$ are defined by (3.7)–(3.9),

respectively, when the competition of A and B is considered.

The properties of the stationary state, where $dn/dt = 0$ and $dz/dt = 0$, may be obtained as follows. For the realizations of $\phi(X)$ mentioned above, Eqs. (3.10) and (3.11) reduce to two polynomials in $\eta \equiv \exp(-2J/k_B T)$. The common stationary solution (n, z) to them may be unstable, however. A necessary condition for stability is

$$\left[\frac{\partial F}{\partial n} \right]_{SS} < 0, \quad (3.12)$$

where SS stands for *stationary state*. Consequently, there is a *limit of metastability* (or *spinodal line*) located at $(\partial F/\partial n)_{SS} = 0$. Given that (3.12) is not sufficient, we eventually need to solve certain ambiguities below by integrating Eqs. (3.10) and (3.11) for several initial conditions (n_0, z_0) and monitoring the actual time evolution.

Consider $\phi(X)$, $\psi(X)$, and (p, T') given. For T high enough, the stationary state is completely disordered, so that it may be characterized by $m = 0$ ($n = \frac{1}{2}$); consequently, $n = v$, $z = w$, and $F(\frac{1}{2}, z; T) = 0$. Thus, the only condition to obtain the stationary state at high T is

$$(1-p)G(\frac{1}{2}, z; T) + pK(\frac{1}{2}, z; T') = 0, \quad (3.13)$$

which determines z for given T , and stability requires $(\partial F/\partial n)_{n=1/2, z} < 0$. The breakdown of the latter marks the onset of a phase transition. A second-order transition corresponds to a critical temperature, say $T_C \equiv T_1 = T_1(p, T')$, where T_1 locates the *spinodal line*, which is the solution of $(\partial F/\partial n)_{n=1/2, z} = 0$, where z follows from (3.13). Then, the only solution that is locally stable for $T > T_C$ is $m = 0$, and stable solutions $m \neq 0$ exist for $T < T_C$. When the transition is of first order, T_1 is the lower limit of local stability for the $m = 0$ solution. The latter is unstable for $T < T_1$, but $m \neq 0$ solutions may exist for $T > T_1$ that are locally stable. Consequently, one defines the *upper limit of local stability* (for $m \neq 0$ solutions), say $T_2 = T_2(p, T')$, as the solution of $(\partial F/\partial n)_{n \neq 1/2, z} = 0$, where (n, z) is the stationary solution of Eqs. (3.10) and (3.11). Summing up, $m = 0$ is the only stable solution for $T > T_2$, while two stationary, locally stable solutions exist for $T_1 < T < T_2$, which correspond to ordered and disordered states, respectively. The time integration of (3.10) and (3.11) leads to one of the two solutions, depending on the initial condition chosen. The method gives no information on which one of them is globally stable, but locates a region of *metastable behavior* between T_1 and T_2 .

Some specific information for any d may be obtained analytically now. The (mean-field) equilibrium results follow when $p = 0, 1$. For $p = 0$, (3.13) reduces to $G(\frac{1}{2}, z; T) = 0$, which leads to $(2z)^{-1} - 1 = \eta$ when detailed balance, $\phi(X) = \eta^{2X} \phi(-X)$, is used. After combining with $(\partial F/\partial n)_{n=1/2, z} = 0$, one gets $z = d[2(2d-1)]^{-1}$; consequently,

$$T_C = 2J/k_B \ln[d(d-1)^{-1}], \quad (3.14)$$

which is the Bethe critical temperature. Moreover, Eqs.

(3.10) and (3.11) reproduce the ordered states of the Bethe theory for $T < T_C$. On the other hand, $p = 1$ implies $K(n, z; T') = 0$ and $n = \text{const}$, which may be seen to imply $v^2 = \xi^2 zw$. That is, the energy is

$$\begin{aligned} e &= [\Lambda + \xi(2m^2 - 1)](\Lambda + \xi)^{-1}, \\ \Lambda &\equiv [1 - m^2(1 - \xi^2)]^{1/2}, \end{aligned} \quad (3.15)$$

which corresponds to the Bethe-Peierls solution for given m and T' . Note that (3.15) is independent of transition rates, as expected for equilibrium states, and of system dimension, which is a feature of the pair approximation. Note also that making $T = T'$ leads to the equilibrium solutions found above for $p = 0$. Namely, $p = 0$ implies $v^2 = \xi^2 zw$, which vanishes the diffusion term $K(n, z; T)$ (with T' replaced by T).

More interesting is the limit $p \rightarrow 1$. According to Eqs. (3.10) and (3.11), the time scale for variations of m (equivalently, n) induced by the reaction process is of order $(1 - p)^{-1}$, which diverges as $p \rightarrow 1$. Contrary to such a slow relaxation of m , the energy e (equivalently, z) changes very rapidly within that limit due to the (fast) diffusion process, whose time scale is of the order of unity. More explicitly, the slow variation of m allows for the diffusion process to stabilize the system at temperature T' before any significant change of m occurs. This produces at each time a condition of *local equilibrium* at temperature T' that is described by (3.15), i.e., $v^2 = \xi^2 zw$, in the present approximation. The stationary states follow from the conditions $F(n, z; T) = 0$ and $K(n, z; T') = 0$. That is, they are also local equilibrium states (3.15) with no dependence on the diffusion rate $\psi(X)$. Then, $T_1 = T_1(p \rightarrow 1, T')$ follows from

$$\sum_{i=0}^{2d} \binom{2d}{i} \xi^i [i - 1 - (2d - i)\xi] \phi(d - i) = 0, \quad (3.16)$$

which has a strong dependence on both ϕ and d , as discussed below. Note that one gets $z = [2(1 + \xi)]^{-1}$ for $m = 0$.

IV. THE ONE-DIMENSIONAL SYSTEM

We first note that (3.9) leads to $K(n, z; T') = \psi(-1)v(v^2 - \xi^2 zw)(n\nu)^{-3}$ for $d = 1$. It implies, in particular, that the existence of a transition does not depend on the diffusion rate. Different realizations of ψ only modify the relative speed of the diffusion process, i.e., they produce a different *effective* value for p , according to the last term in Eq. (3.11). On the other hand, we get from (3.13) and $(\partial F / \partial n)_{n=1/2, z} = 0$ the following information: (1) A critical point exists at $T_C = 0$ for any value of p and T' if $\phi(X) = (1 + e^X)^{-1}$. (2) The zero- T critical point washes out for $\phi(X) = \min\{1, e^{-X}\}$, unless at least one of the two conditions $T' = 0$ or $p = 0$ holds; otherwise, only $m = \pm 1$ solutions occur for $T = 0$. (3) When either $\phi(X) = e^{-(1/2)X}[\cosh(\beta J)]^2$ or $\phi(X) = e^{-(1/2)X}$, a surface of critical points exists, $T_C = T_C(p, T')$, which is the solution of

$$\begin{aligned} 4\alpha\Gamma^2(1 - \Gamma)^2[(2\Gamma\xi + \Gamma - \xi)(2\Gamma\xi - \Gamma - \xi)]^{-1} \\ = p\psi(-1)(1 - p)^{-1}, \end{aligned} \quad (4.1)$$

where

$$\begin{aligned} \alpha &\equiv \Gamma^{-1} \text{ for } \phi(X) = e^{-(1/2)X} \\ &\equiv 4(1 + \Gamma)^{-2} \text{ for } \phi(X) = e^{-(1/2)X} \cosh(\beta J), \end{aligned} \quad (4.2)$$

and $\Gamma \equiv \exp(-2J/k_B T_C)$. Figure 9 illustrates the solutions of (4.1) for the second choice of α in (4.2) and two different diffusion rates, namely the Metropolis and Kawasaki ones.

Two important facts are revealed in Fig. 9. On the one hand, the form of ψ becomes irrelevant for the limiting cases, $T' = 0, \infty$ and $p = 0, 1$, and it induces only minor quantitative changes otherwise. On the other hand, the present mean-field treatment for $T' = \infty$ leads to the transition points obtained previously by the Monte Carlo method [10]. This agreement deserves a comment, however. The computer simulation of a chain with 10^4 sites suggested the existence of first-order transitions for $p > 0.5$, and perhaps also for any $p > 0$ (i.e., the one-dimensional system revealed a tendency towards phase segregation that only becomes quite clear in the MC experiments for $p > 0.5$), while the transitions implied by (4.1) are always of second order. One is inclined to believe that the apparent MC discontinuities are not real. In fact, the system typically undergoes a very slow monotonic decay during the MC relaxation, rather than evolving through actual metastable states (as one would expect in a first-order transition), and it is exactly known that the transition is of second order for $p \rightarrow 1$. Nevertheless,

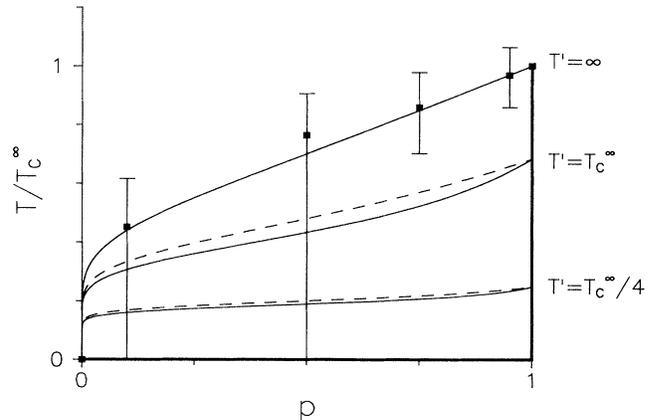


FIG. 9. Phase diagram for a one-dimensional system evolving by reaction rates *à la* De Masi. The horizontal line corresponds to the equilibrium zero- T critical point, where $p < 1$ is a trivial parameter. The rightmost vertical line represents a line of critical points for $p \rightarrow 1$ and any $T' > T$. The solid and dashed curves correspond, respectively, to diffusion rates *à la* Metropolis and *à la* Kawasaki for different values of T' , as indicated. The symbols represent Monte Carlo data [10] whose error bars (which are consistent with having a phase transition only for $p > 0.5$) are also shown. $T_C^\infty \equiv 2J(k_B \ln 3)^{-1}$ is the exact critical temperature for $p \rightarrow 1$, $T' = \infty$.

we still trust the MC evidence [10] of long-range order (in particular, the lattice was large enough so that crucial finite-size effects should be absent). Summing up, we believe that the system is described accurately by our mean-field solution, at least for $p \geq 0.5$. This seems confirmed by Fig. 10, describing a qualitative agreement between mean-field and MC results for m and for C as defined in (2.1) and (2.3), respectively, at least for $p \geq 0.5$. Two more facts support our conclusion: (1) The present mean-field theory reproduces well the fact, revealed by both exact and MC results, that the one-dimensional system has no phase transition at finite T for Metropolis and Kawasaki reaction rates, for instance; (2) Eq. (4.1) reduces, for $p \rightarrow 1$, to $\Gamma = \zeta(1+2\zeta)^{-1}$, which equals the corresponding exact solution for arbitrary T' [10].

The reason for the agreement between mean-field and exact theories is that our method of solution is independent of the size of the involved cluster D for $d=1$, so that the only approximation is our restriction to pair correlations, which are actually the only ones when local equilibrium sets in for $p \rightarrow 1$. The latter assertion follows from the next argument. Assuming homogeneity (which is implicit in the method), the exact equation for the magnetization when $d=1$ and $p \rightarrow 1$ is

$$\frac{d\langle s_x \rangle}{dt} = -2 \sum_s s_x \phi(\delta H) \mu_{\text{eq}}^{T'}(\mathbf{s}; m); \quad (4.3)$$

cf. Eq. (1.2). Summing here over $\mathbf{s} - \mathbf{s}_D$, where \mathbf{s}_D is any configuration of D that involves s_x and its two nearest neighbors, one gets

$$\frac{d\langle s_x \rangle}{d\tau} = -2 \sum_{\mathbf{s}_D} s_x Q_D(\mathbf{s}_D) \phi(\delta H), \quad (4.4)$$

where one has, for $d=1$, that

$$Q_D(\mathbf{s}_D) = \sum_{\mathbf{s} - \mathbf{s}_D} \mu_{\text{eq}}^{T'}(\mathbf{s}; m) = p(s_x) p(s_{x-1} | s_x) p(s_{x+1} | s_x). \quad (4.5)$$

Now, the result (4.4) and (4.5) may be organized as in (3.5), i.e.,

$$\frac{d\langle s_x \rangle}{dt} = -2 \sum_{s_x = \pm 1} \sum_{i=0}^2 s_x Q_D(s_x, i) \phi[s_x(i-1)], \quad (4.6)$$

where $Q_D(s_x, i)$ is defined in (3.6). When one takes $n = \frac{1}{2}(1 + \langle s_x \rangle)$ here, it follows (3.7), which (together with $v^2 = \zeta^2 z w$) is the solution of our method, i.e., the latter produces indeed the exact solution for $p \rightarrow 1$. This argument also suggests that one might expect the same result to be valid, at least approximately, for a range of p values smaller than unity. It is clear, however, that, lacking more general exact results, additional MC data is needed to conclude more definitely about the problems suggested here.

V. THE TWO-DIMENSIONAL SYSTEM

Conditions (3.12) and $(\partial F / \partial n)_{n=1/2, z} = 0$ lead, respectively, to

$$\begin{aligned} \phi(-2)(\Gamma^4 - u^4) + 2\phi(-1)u(\Gamma^2 - u^2) \\ = 6p(1-p)^{-1}zu(u - \zeta)[1 - 2(1 - \zeta)z]\alpha \end{aligned} \quad (5.1)$$

and

$$\begin{aligned} \phi(-2)[(3z - 2)\Gamma^4 + 3u^4z] - 2\phi(-1) \\ \times [6u^2z\Gamma^2 + u^3(1 - 6z)] - 6\phi(0)u^2(1 - 3z) = 0. \end{aligned} \quad (5.2)$$

Here

$$\begin{aligned} \alpha \equiv \psi(-3)(u^4 + \zeta^2 u^2 + \zeta^4) \\ + \psi(-2)4zu(u^2 + \zeta^2) + \psi(-1)5u^2z \end{aligned} \quad (5.3)$$

contains any dependence on the diffusion rate; $u \equiv (2z)^{-1} - 1$, and $\Gamma \equiv \exp(-2J/k_B T_1)$.

Equations (5.1) and (5.2) transform into two polynomials of Γ for the above choices of ϕ . In particular, they are polynomials of second order in Γ^2 for the Metropolis reaction rate, and one may convince oneself that they reduce to the system of equations studied by Dickman [8] when $T' = \infty$. For $p=0$, (5.2) leads to $\Gamma = u$, which produces the Bethe critical temperature $\Gamma = \frac{1}{2}$ when used in Eq. (5.1), independently of the rates, as expected. For $p \rightarrow 1$, (5.2) leads to $\zeta = u$, which produces

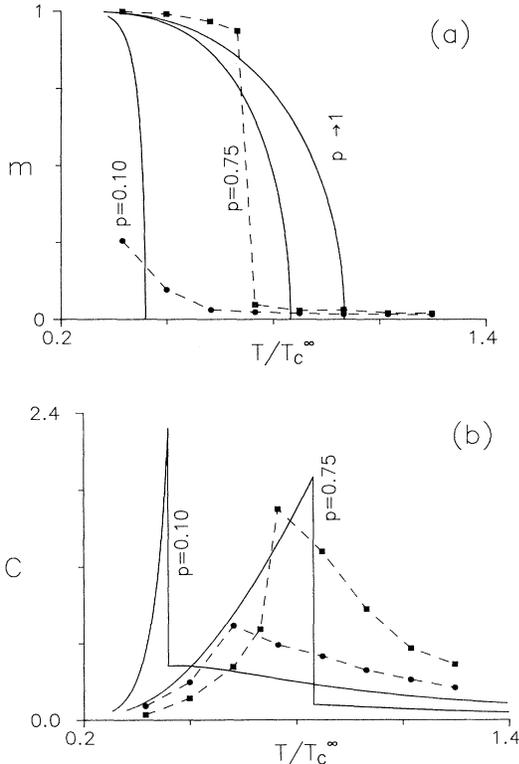


FIG. 10. (a) Order parameter defined in (2.1), and (b) the energy fluctuations defined in (2.3), both for $d=1$, $T' = \infty$, and reaction rates *à la* De Masi. The solid lines are the mean-field result for different values of p , as indicated. The symbols correspond to MC data [10] (squares for $p=0.75$, circles are for $p=0.10$); the dashed lines are a guide to the eye.

$$\begin{aligned} \phi(-2)[3\xi^4 - (1+4\xi)\Gamma^4] - \phi(-1)4[3\xi^2\Gamma^2 + \xi^3(\xi-2)] \\ + \phi(0)6\xi^2(1-2\xi) = 0 \end{aligned} \quad (5.4)$$

when used in (5.1). Equation (5.4), which corresponds to (3.16) for $d=2$, gives $T_1 = T_1(T')$ for different ϕ 's. In particular, one has for the Metropolis rate that

$$\Gamma^2 = \frac{\xi(6+20\xi+19\xi^2-4\xi^3)^{1/2}-6\xi^2}{1+4\xi}, \quad (5.5)$$

which reduces to the known solution $\Gamma^2=0.081$ for $T'=\infty$.

Under condition $T'=\infty$, the limit $p \rightarrow 1$ reproduces the known exact solutions, as described for $d=1$ above. The reason is that $T'=\infty$ induces local equilibrium, where most correlations are suppressed, which makes the pair approximation exact and our method independent of D . Thus, the only actual restriction in our description is the implicit assumption about homogeneity. In particular, we obtain a transition of second order when the reaction rate is implemented by either Kawasaki or van Beijeren-Schulmann rates; the corresponding critical point is located at $J/k_B T_C = 0.3236$ and 0.2554 , respectively. The transition is of first order when the reaction rate is Metropolis, however. The main equation for the latter case is

$$\begin{aligned} m^{-1}(dm/dt) = (5+10m^2+m^4)\Gamma^4 + 4(3-2m^2-4m^4)\Gamma^2 \\ - (1+2m^2-3m^4), \end{aligned} \quad (5.6)$$

whose solutions are of the form

$$m = \begin{cases} m^* & \text{if } T < T_1 \\ m^*, 0 & \text{if } T_1 < T < T_2 \\ 0 & \text{if } T > T_2. \end{cases} \quad (5.7)$$

Here T_1 and T_2 are, respectively, the solution of $\Gamma^2=0.081$ and $5\Gamma_2^6-3\Gamma_2^4+7\Gamma_2^2-1=0$; $\Gamma_2 \equiv \exp(-2J/k_B T_2)$. A transition temperature may be defined as $T_{tr} \equiv \frac{1}{2}(T_1+T_2)$.

The situation under more general conditions may be very varied, e.g., both first- and second-order transitions, which correspond to $T_1 \neq T_2$ and $T_C = T_1 = T_2$, respectively, may occur for a given rate. A systematic study may proceed by computing $m = m(T)$ and $e = e(T)$ for selected choices of p , T' , $\phi(X)$, and $\psi(X)$, as described in Sec. III. The ensuing picture when both rates are implemented by the Metropolis algorithm may be summarized as follows (cf. Figs. 11 and 12).

(1) $T_1(p, T')$ exists for any pair of values (p, T') such that the system exhibits $m \neq 0$ solutions for $T < T_1$.

(2) A sort of essential asymmetry around the Bethe critical temperature T_C^B exists. Namely, the critical value of z is always larger than $\frac{1}{3}$ for any p (only) when $T' < T_C^B$, as depicted by Fig. 13. We interpret this as an evidence (in fact, the only one that is allowed by our method) that inhomogeneous solutions with $m=0$ then exist due to a tendency of the diffusion mechanism to induce phase segregation.

(3) One may distinguish up to three qualitatively

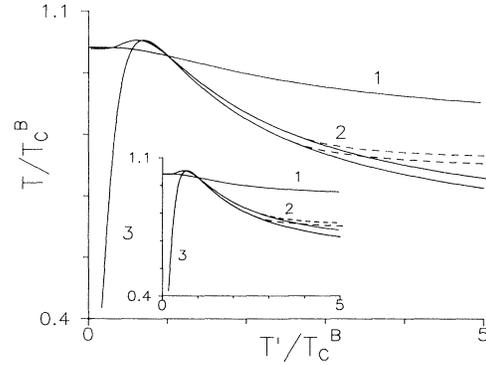


FIG. 11. Phase diagram for the two-dimensional system as implied by the theory in Sec. V. The solid lines represent $T_1(p, T')$; the dashed lines represent $T_2(p, T')$. The transition is of first order when $T_1 \neq T_2$, and of second order otherwise. $T_C^B \equiv 2J(k_B \ln 2)^{-1}$ is the equilibrium Bethe critical temperature. The main graph corresponds to a system evolving by Metropolis reaction and diffusion rates, and different values of p : curves 1, 2, and 3 are for $p=0.5$, $p=0.95$, and $p \rightarrow 1$, respectively. The inset, exhibiting the same behavior, corresponds to a Kawasaki diffusion rate.

different types of behavior for $p \rightarrow 1$ (cf. curves 3 in the main graph of Fig. 11 and in the inset of Fig. 12): (I) transitions are equilibriumlike, second order for $T_C^B < T' < 2.574T_C^B$; (II) transitions are of first order for $T' > 2.574T_C^B$; (III) while second-order transition occur for $T' < T_C^B$, the situation is more subtle than for case I. Namely, the solutions are inhomogeneous (as described above) near $T_C(T')$, and the following facts are observed for very small values of T' , say for $T' < 0.1T_C^B$: (i) The variations of both m and e with T are rather strong near $T_C(T')$, though the latter are typically an order of magnitude smaller than the former. (ii) The numerical treatment of Eqs. (3.12) and (3.13) for different initial condi-

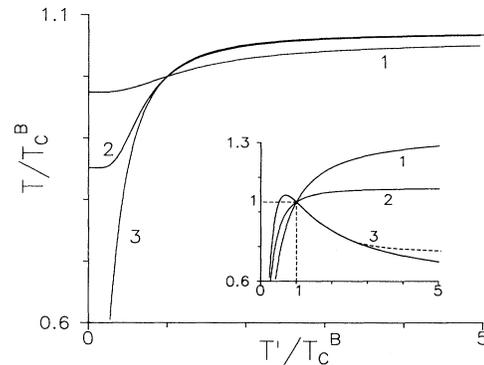


FIG. 12. Main graph here represents the same situation as in the main graph of Fig. 11, but for a reaction rate of the Kawasaki type. The behavior (not shown) is the same when both reaction and diffusion rates are of the Kawasaki type. The inset corresponds to $p \rightarrow 1$ for $\psi(X) = \min\{1, e^{-X}\}$ and different reaction rates: curve 1 corresponds to $\phi(X) = e^{-(1/2)X}$, curve 2 is for $\phi(X) = [1 + e^X]^{-1}$, and curve 3 is for $\phi(X) = \min\{1, e^{-X}\}$.

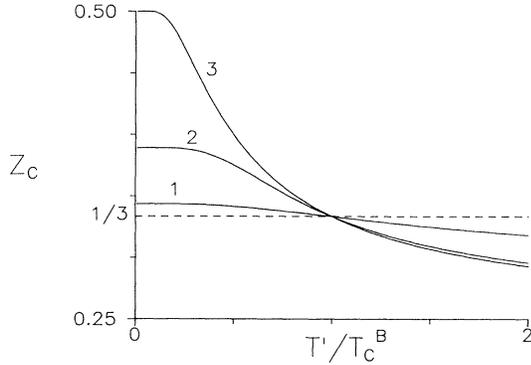


FIG. 13. Probability of NN_{++} pairs, z , at critical temperature $T=T_1(T')$ for different values of p (curves 1, 2, and 3 are for $p=0.5$, $p=0.95$, and $p\rightarrow 1$, respectively) when reaction and diffusion rates are implemented by the Metropolis algorithm. This illustrates the asymmetry around $T'=T_c^B$, which is discussed in the main text.

tions reveals very slow relaxations. This is apparently due to the existence of a full phase segregation, which makes the system rather insensible to attempted changes $s_x \rightarrow -s_x$ at low T' . The behavior III, where $T_c(T') \rightarrow 0$ as $T' \rightarrow 0$, is the one that manifests itself as *type II'* in a MC experiment; cf. Sec. II. In any case (i.e., for I, II, or III), z and $\sigma = \frac{1}{4}z\omega v^{-2}$ depend on T' but are independent of T , given that any short-range order is dominated by the diffusion process when $p \rightarrow 1$.

(4) The behavior for $p=0.95$ (cf. curve 2 in the main graph of Fig. 11) is qualitatively similar to the one for $p\rightarrow 1$, except for the following facts: (i) The *tricritical point* separating cases I and II is located now at $T'=2.622T_c^B$. (ii) There is no indication of a behavior such as III (which is still evident in our equations for $p=0.999$ and $T'=0.05T_c^B$), and there is some decreasing of $T_c(T')$, but not a tendency towards zero, as $T' \rightarrow 0$. Both z and σ depend on T ; e.g., $\sigma(T)$ has a monotonic decrease with T and presents a discontinuity for case II.

(5) As p is further decreased, the region corresponding to case I tends to occupy a larger range of T' values (e.g., the tricritical point for $p=\frac{1}{2}$ occurs at $8.152T_c^B$, and no first-order transition occurs for $p < 0.364$), and $T_1(T')$ decreases monotonically with T' ; cf. curve 1 in the main graph of Fig. 11.

In general, the steady state is observed to depend qualitatively on $\phi(X)$. This is evident in the inset of Fig. 12, where the three curves correspond to different relations of $\phi(X)$; the main graph of Fig. 12 illustrates the phase diagram for varying values of p when the reaction rate is implemented by the Kawasaki algorithm. The main results for this case may be summarized as follows: (i) The transitions are always of second order, with $T_c = T_1(T')$ increasing monotonically with T' . (ii) The situation for $T' = \infty$ is qualitatively similar to the one described in a MC experiment [15] for a Glauber reaction rate; in particular, T_c increases monotonically with p . The Glauber rate, which was originally defined for $d=1$, cannot be

written for $d=2$ in the form (3.1) (consequently, it cannot be included in our analysis); it has an effect on dynamics that is close to the one by the Kawasaki rate, however. (iii) When $p\rightarrow 1$, the short-range order is determined by diffusion at temperature T' , and one may distinguish two cases: the behavior is equilibriumlike of type I for any $T' \geq T_c^B/10$, approximately, while it is of type III (as for Metropolis rates above) when $T' \leq T_c^B/10$. (iv) The behavior is always equilibriumlike of type I for any $p < 1$.

The investigation of different realizations for $\psi(X)$ is also interesting. The inset in Fig. 11 illustrates the case of Metropolis reaction and Kawasaki diffusion rates (which is to be compared with the main graph). We obtain the same qualitative behavior as in the main graph of Fig. 12 when both rates are of the Kawasaki type. A main conclusion here is that, independently of p , different choices for the diffusion rate only seem to induce minor quantitative changes, unlike the case for the reaction rate.

VI. DISCUSSION

The *reaction-diffusion lattice gas*, whose kinetics consist of a competition between creation-annihilation (*reaction*) and diffusion processes at two different temperatures, is one of the most interesting microscopic models at hand in which nonequilibrium situations occur. In addition to the fact that macroscopic, hydrodynamiclike equations have been derived rigorously for it [5–7], it exhibits a variety of ordering phenomena as one modifies the system parameters, and models some actual situations in nature [4]. The Monte Carlo (MC) simulation and mean-field analysis reported in the present paper add up to a series of recent exact, mean-field, and MC results (cf. Refs. [4–15], and references therein). A brief description of the emerging picture concerning nonequilibrium phase transitions follows; eventually, some specific questions are discussed.

The influence on system properties of the reaction and diffusion processes (which are governed by rates $(1-p)\phi(X)$ and $p\psi(X)$, where X represents the corresponding energy cost, $[H(s')-H(s)]/k_B\Omega$, with $\Omega=T$ or T' , respectively) is uneven. Exact [10] and mean-field theory reveal that different realizations of $\psi(X)$ induce the same qualitative behavior with some minor quantitative differences, e.g., different *effective* values for p . In any case, $\psi(X)$ is irrelevant for $T'=0, \infty$, as expected. The choice of $\phi(X)$ has a decisive influence on the nature of the stationary state, however. In particular, the one-dimensional system has long-range order below $T_c(p, T') > 0$ only for a certain family of reaction rates that includes $\phi(X) = 1 - \alpha s_x [s_{x-1} + s_{x+1}] + \alpha^2 s_{x-1} s_{x+1}$, $\alpha \equiv \tanh(J/k_B T)$ [5]. This family has in common that rates favor low-energy states rather atypically, in which the Metropolis algorithm is excluded. The latter and other choices induce a zero- T critical point instead, if any; e.g., the zero- T critical point is washed out except if either $T'=0$ or $p=0$ occurs, and only $m = \pm 1$ solutions arise otherwise, in the Metropolis case. It also follows that correlations for $d=1$ decay ex-

ponentially with distance, in agreement with some expectations [21]; cf. Ref. [10] also.

The present mean-field approach suggests that the phase transition that occurs for $d = 1$ at $T_C(p, T') > 0$ is of second order, in agreement with exact results for a completely random and fast diffusion (i.e., $T' = \infty$ and $p \rightarrow 1$). In addition, the latter predicts classical behavior for the order parameter, namely $m = \alpha^{-1}(2\alpha - 1)^{1/2} > 0$ for $\alpha > \alpha_C = \frac{1}{2}$. This is essentially similar to the result obtained for a generalization [10] of the Glauber rates [1] to $d = 2$; thus, one may argue that the effect of the atypical rates mentioned in the preceding paragraph, as compared to the Glauber ones, is to produce an effective increase to $d + 1$ of the actual dimension of the system. It is suggested by mean-field theory that long-range order occurs at finite T' for any $p > 0$. Previous MC simulations for $T' = \infty$ indicated that the transition may occur for $p \gtrsim \frac{1}{2}$ only, and apparent discontinuities were observed (cf. Figs. 9 and 10); the computer study of the one-dimensional case is relatively difficult, however, and the latter observation may be an artifact due to very slow relaxations. It would be interesting to perform further numerical studies of this and related cases, in particular, to compare our equations in Sec. IV with MC data for $d = 1$, trying to conclude more definitely about the nature of the phase transition as one varies p , its relation to the classical one occurring for $p \rightarrow 1$, and the possible existence of discontinuities and/or a tricritical point.

As expected, mean-field theory is an accurate description of many properties of the reaction-diffusion lattice gas. The following situation is revealed by mean-field theory for $p \rightarrow 1$, in agreement with exact results. Reaction induces a slow relaxation of m , whose time scale is of order $(1-p)^{-1}$, while diffusion produces very rapid changes of the energy, whose time scale is of order p^{-1} . Consequently, a condition of *local equilibrium* (where most correlations are suppressed) at temperature T' sets in. On the other hand, the mean-field method is naturally independent of the size of the involved cluster D as long as $d = 1$, so that the only approximation that is implicated is a restriction to pair correlations that are exact at local equilibrium. We thus get for $d = 1$ perfect agreement with the exact solution for arbitrary T' [10]. Such an agreement occurs also for $d = 2$ when $T' = \infty$, which induces local equilibrium as well.

The two-dimensional system is rather well understood now. The analytical and MC studies describe a phase transition whose nature may vary, depending both on $\phi(X)$ and p , from second to first order as T' is increased. For example, the following types of behavior arise in MC experiments when both reaction and diffusion mechanisms are implemented by the Metropolis algorithm: For $p = 0.95$, the transition is of second order insofar as $T_C^0 < T' < 5.35T_C^0$ (where T_C^0 is the Onsager critical temperature), the transition is standard of first order when $T' > 5.35T_C^0$, and it has weak discontinuities and some peculiarities, such as very long-lived metastable states

and very slow evolutions, when $T' < 0.8T_C^0$; i.e., the temperature of the (fast) diffusion process is crucial. The speed of the diffusion process is also crucial: the regions in which two phases coexist tend to disappear as p is decreased, and only continuous transitions occur for $p < 0.83$. The mean-field description (which agrees with the exact solution for $p \rightarrow 1$ and $T' = \infty$ [8] only) reveals the same qualitative picture; the third mentioned class of behavior shows up then as a second-order phase transition with inhomogeneous states as $p \rightarrow 1$ (but not for $p < 1$), however. Further studies of the coexistence of phases that is suggested by both MC and mean-field results would be interesting. On the other hand, a diffusion Kawasaki rate does not modify qualitatively the steady state (within a mean-field description), while the use of the same rate for the reaction mechanism induces an important change: no standard first-order phase transitions exist anymore, but second-order ones with T_C an increasing function of both p and T' do exist.

A finite-size study of the MC data for Metropolis rates reveals that the critical behavior is always indistinguishable from the equilibrium, Onsager one. It is consistent with exact results for $T' = \infty$ and $p \rightarrow 1$, indicating a phase transition of first order when $\phi(X)$ is of the Metropolis type, and of second order otherwise. The latter is classical (e.g., $\beta = \frac{1}{2}$), however, given that $T' = \infty$ and, more effectively, $p \rightarrow 1$ induce mean-field behavior. (Mean-field behavior has been shown to be a consequence of infinite-range diffusion for any $p \leq 1$ [13] as well.) The limit $p \rightarrow 1$ is singular, however: a relatively slower process with $T' = \infty$ induces for Metropolis rates the existence of a tricritical point at $p \approx 0.83$, so that the phase transition is of first order for $p > 0.83$ only. No similar systematic study seems to exist in the literature for other choices of $\phi(X)$, except some limited (MC) data suggesting that no first-order transition occurs for Glauber rates when $p < 1$ [15], in agreement with the exact result for $p \rightarrow 1$ [9,10]. It would be interesting to conclude with confidence about possible changes of the second-order phase transitions as one decreases p from the fast diffusion limit, given that one might expect there a changeover from classical to equilibrium critical behavior. It would also be interesting to study the derivation of macroscopic equations and transport coefficients for finite T' following the previous trends for $T' = \infty$ [5]. Finally, it may be remarked that the complex behavior of the reaction-diffusion lattice gas does not seem to fit the simple picture in Ref. [19] for some nonequilibrium lattice systems.

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- [1] R. J. Glauber, *J. Math. Phys.* **4**, 294 (1963).
- [2] K. Kawasaki, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, London, 1972), Vol. 4.
- [3] P. L. Garrido and J. Marro, *Phys. Rev. Lett.* **62**, 1929 (1989).
- [4] See, for instance, D. G. Aronson and H. F. Weinberger, *Partial Differential Equations and Related Topics* (Springer-Verlag, Berlin, 1975); H. Haken, *Synergetics* (Springer-Verlag, Berlin, 1978), and references therein.
- [5] A. De Masi, P. A. Ferrari, and J. L. Lebowitz, *J. Stat. Phys.* **44**, 589 (1986).
- [6] J. L. Lebowitz, *Physica A* **140**, 232 (1986).
- [7] J. L. Lebowitz, E. Presutti, and H. Spohn, *J. Stat. Phys.* **51**, 841 (1988).
- [8] R. Dickman, *Phys. Lett. A* **122**, 463 (1987).
- [9] J. M. González-Miranda, P. L. Garrido, J. Marro, and J. L. Lebowitz, *Phys. Rev. Lett.* **59**, 1934 (1987).
- [10] P. L. Garrido, J. Marro, and J. M. González-Miranda, *Phys. Rev. A* **40**, 5802 (1989); J. J. Alonso *et al.* (unpublished).
- [11] P. Bellon and G. Martin, *Phys. Rev. B* **39**, 2403 (1989).
- [12] M. Droz, Z. Rácz, and J. Schmidt, *Phys. Rev. A* **39**, 2141 (1989).
- [13] M. Droz, Z. Rácz, and P. Tartaglia, *Phys. Rev. A* **41**, 6621 (1990); *Physica A* **177**, 401 (1991).
- [14] J. M. González-Miranda and J. Marro, *J. Stat. Phys.* **61**, 1283 (1990).
- [15] J.-S. Wang and J. L. Lebowitz, *J. Stat. Phys.* **51**, 893 (1988).
- [16] N. Metropolis, A. W. Rosenbluth, M. M. Rosenbluth, A. H. Teller, and E. Teller, *J. Chem. Phys.* **21**, 1087 (1953).
- [17] J. Marro, P. L. Garrido, A. Labarta, and R. Toral, *J. Phys. Condens. Matter* **1**, 8147 (1989).
- [18] A. Labarta, J. Marro, and J. Tejada, *Physica B* **142**, 31 (1986).
- [19] G. Grinstein, C. Jayaprakash, and Y. He, *Phys. Rev. Lett.* **55**, 2527 (1985).
- [20] M. E. Fisher and M. N. Barber, *Phys. Rev. Lett.* **28**, 1516 (1972); D. P. Landau, *Phys. Rev. B* **14**, 255 (1976).
- [21] Z. Cheng, P. L. Garrido, J. L. Lebowitz, and J. L. Vallés, *Europhys. Lett.* **14**, 507 (1991), and references therein.
- [22] P. L. Garrido, J. Marro, and R. Dickman, *Ann. Phys. (N.Y.)* **199**, 366 (1990).
- [23] H. van Beijeren and L. S. Schulmann, *Phys. Rev. Lett.* **53**, 806 (1984).