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Universality and diffusion in nonequilibrium critical phenomena

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Nonequilibrium lattice models in which particles are produced autocatalytically, and annihilated spontaneously, are studied via simulations, series expansions, and mean-field theory. There is a continuous transition to an absorbing state as the creation rate is reduced below a critical value. The results demonstrate universality of critical behavior under variations in reaction scheme. Models with binary and triplet annihilation processes are also considered. In the latter case, competition between diffusion and annihilation yields a surprising new phase at low creation rates. For sufficiently rapid diffusion there is a nonabsorbing steady state for any nonzero creation reate.

I. INTRODUCTION

Consider a many-particle system in a macroscopically steady nonequilibrium state. As certain "control parameters" (e.g., the values of the imposed temperature or density gradients, chemical feed rates, etc.) are varied, such a system may exhibit a nonequilibrium phase transition (NEPT). In analogy with equilibrium thermodynamics, a NEPT denotes a singular dependence of steady-state properties upon the control parameters, marking a transition between distinct regimes of operation. NEPT's are often associated with the appearance of new levels of organization (e.g., rolls at the onset of convection or oscillations and waves in chemical reactions) and so are currently of great interest in physics and biology.[1,2] Continuous NEPT's or *nonequilibrium critical points* (NECP's) exhibit many features associated with equilibrium critical phenomena: long-range correlations, a well-defined order parameter, and singularities characterized by critical exponents. NECP's are of interest in condensed-matter physics, since they form a new and largely unexplored domain for critical phenomena. A number of models are presently being studied in efforts to understand specific NECP's. Examples are surface-reaction models[3,4] which describe the poisoning of a catalyst and the driven lattice gas pertinent to ionic conductors.[5]

The present work is motivated by the question of universality in NECP's. While the factors influencing nonequilibrium critical behavior are in general not well understood, this issue has been examined recently for a class of stochastic models in which particles are created autocatalytically and spontaneously disappear. The critical point in such models marks a transition to an *adsorbing state,* devoid of particles. A simple example is Schlögl's "first model"[6] which describes an autocatalytic system in which particles (X) participate in the reactions $X \rightarrow 0$, $X \rightarrow 2X$, and $2X \rightarrow X$ at specified rates. For sufficiently large creation rates, it is possible to maintain an active steady state (i.e., with a nonzero population of particles); as the creation rate η is decreased there is a continuous transition to the absorbing state, with the order parameter (the particle density ρ), decaying asymptotically as $\rho \propto (\eta - \eta_c)^{\beta}$.

In Schlögl's original formulation—a mean-field theory (MFT) without spatial dependence—the exponent β is 1 for all dimensions d. When the model is given spatial structure, either on a lattice or in continuous space (with reactions proceeding *locally)* the critical exponents assume non-mean-field values, which depend on d. Early investigations, pioneered by Grassberger and de la Torre[7] and Janssen[8] were spurred by a correspondence between the chemical kinetic model and Reggeon field theory (RFT).[9] It transpired that the critical behavior of Schlögl-RFT-type models (in d spatial dimensions) is the same as for directed percolation[10] (in $d+1$ dimensions, with the oriented dimension corresponding to time in the Schlögl-RFT models). This type of critical behavior (which will be called RFT in the following) is by now rather well understood: The upper critical dimension is $d_c = 4$, and critical exponents have been estimated to good precision. In particular, for $d=1$ the order-parameter critical exponent is $\beta = 0.277 \pm 0.001$.[9,11]

Early studies[7] revealed a degree of robustness for RFT critical behavior, showing that critical exponents are not altered by variations in the relative probabilities of single- and double-particle production, on a lattice where sites may be at most doubly occupied. Schlogl's *second* model[6] (i.e., the reactions $2X \rightleftharpoons 3X$, $X \rightleftharpoons 0$, at given rates) was expected (on the basis of simple mean-field theory) to exhibit a *discontinuous* transition over a range of parameter values. Simulations[12] instead revealed a RFT critical point, leading Grassberger to conjecture that all single-component models with a unique absorbing state belong to the RFT universality class. This assertion is supported by field-theoretic arguments[8,12,13] which predict that a continuous transition to an absorbing state will be of the RFT type, unless the (renormalized) rate for the reaction $2X \rightarrow X$ (i.e., the most relevant term in the dynamics) is zero. RFT, therefore, appears to be analogous to the Ising model ϕ^4 field theory in equilibrium as the generic critical behavior associated with a scalar order parameter.

Of course, the field-theoretic prediction of universality ought to be tested for specific models: One would like to verify the robustness of RFT critical behavior for a wider range of kinetic rules than has been explored until now.

One would also like to know the minimal modifications required to produce non-RFT behavior. The results to be presented in this paper support the universality of RFT in a wide range of models. On the other hand, simulations of a model with diffusion and cluster annihilation reveal a new and surprising phase diagram. In the following section, I present simulation- and series-analysis results for several nondiffusive models, typified by the contact process. Diffusive models are considered in Sec. III, which is followed by a summary in Sec. IV.

II. THE CONTACT PROCESS AND RELATED MODELS

The systems considered in this work are stochastic lattice models or *interaction particle systems*[14] characterized by a set of occupation variables σ_i ($i \in \mathbb{Z}^d$, $\sigma_i = 0$, 1 for site i vacant or occupied, respectively). The models evolve via creation and annihilation of particles, subject to local, irreversible Markovian rules which admit a single absorbing state (an empty lattice). Perhaps the simplest such model is the *contact process*[15] (CP), which was introduced as a model for an epidemic, and may be regarded as a realization of Schlögl's first model on a lattice. In the CP each particle generates new particles at rate η. They appear randomly at any of the vacant nearest neighbors of the parent particle, so that the creation rate at a vacant site is $n\eta/2d$, where n is the number of occupied nearest neighbors. Particles disappear at unit rate, independent of the states of other sites. The CP has an active steady state for each $d \geq 1$, for $\eta > \eta_c(d)$.[14,15] Numerical work (simulations and series analysis) indicates that the transition is continuous. The series expansions[9,11] (in one dimension) yield $\eta_c \cong 3.299$ and $\beta \cong 0.277$, the latter reflecting RFT critical behavior.

I have studied several variants of the contact process. The A model, introduced[16] as a simplified model for poisoning of a catalytic surface,[3] differs from the CP solely in that the creation rate at site I is η, provided at least one neighbor of i is occupied. A second variation, called $N3$, has a creation rate of η ($\eta/4$) for sites with two (one) occupied neighbors. These models may be considered in a unified fashion if we introduce a parameter ζ, so that the creation rate at a (vacant) site with two (one) occupied neighbors is η ($\zeta\eta$). Then the A, CP, and $N3$ models correspond to $\zeta = 1$, $\frac{1}{2}$, and $\frac{1}{4}$, respectively. A mean-field treatment at the pair level (following the approach employed in Ref. 4) yields a steady-state density $\bar{\rho} \propto (\eta - \eta_c)$ for $\eta > \eta_c = \zeta^{-1}$, i.e., there is a continuous transition to the absorbing state for all $\zeta > 0$, and $\beta_{\mathrm{MFT}} = 1$. While mean-field theories may provide a qualitatively correct phase diagram,[4,17] they are clearly inadequate for a detailed description of critical behavior, and more quantitative methods must be sought. Series expansions for the steady-state occupation fraction in the one-dimensional CP, A, and $N3$ models have recently been derived via time-independent perturbation theory.[11,16] These rather long series [to 16th order in $v = 1/(1+\eta)$] yield β values which clearly place the models in the RFT universality class (see Table I). It seems very reasonable to expect the same critical behavior for all ζ in the interval $[\frac{1}{4}, 1]$. Given the mean-field result noted above, one might conjecture that there is a RFT-type transition for all $\zeta > 0$.

An important feature of some catalytic surface reactions[3] is pairwise adsorption, and so the relevance of such a kinetic rule to critical behavior is of particular interest. Since the order parameter for the surface reaction model is the *vacancy* density, pairwise adsorption corresponds to pairwise *destruction* of particles in a model like the CP. The *binary annihilation* or $A2$ model features pairwise annihilation (at unit rate) of particles occupying neighboring sites and creation as the CP. The presently available (eight term) series for the binary annihilation model is too short to permit a reliable estimation of β.[11] We, therefore, turn to Monte Carlo simulations.

Simulations of the $A2$ and $N3$ models in one dimension have been used to estimate the steady-state occupation fraction $\bar{\rho}$, following a procedure similar to that employed for the A model.[16] A step in the simulation algorithm consists of (1) choosing the process [creation with probability $\eta/(1+\eta)$, annihilation with probability $1/(1+\eta)$]; (2) choosing a site i at random; (3) performing

TABLE I. Summary of reaction rules and critical parameters for the models considered in this work. Occupied and vacant sites are denoted by solid and open circles, respectively. Creation rates are for the central site of the triplet. The annihilation process is shown in parentheses.

Model	Creation rate ●○○	Creation rate ●○●	Annihilation	η_c	β
CP	$\eta/2$	η	1 (●→○)	3.299[a,b]	0.277(1)[a,b]
A	η	η	1 (●→○)	1.742[b]	0.277(1)[b]
$N3$	$\eta/4$	η	1 (○→○)	6.169[b]	0.278(1)[b]
$A2$	(same as CP)		1 (●●→○○)	5.370(1)	0.27(2)
$D3$	CP rates with $\eta \to \frac{(1-D)\eta}{1+\eta}$		$\frac{1-D}{1+\eta}$(●●●→○○○)		
			$D=0.0$:	6.72(1)	0.28(2)
			$D=0.1$:	4.728(7)	0.30(2)
			$D=0.5$:	0.718(1)	0.28(2)

[a]Reference 9.
[b]Reference 10.

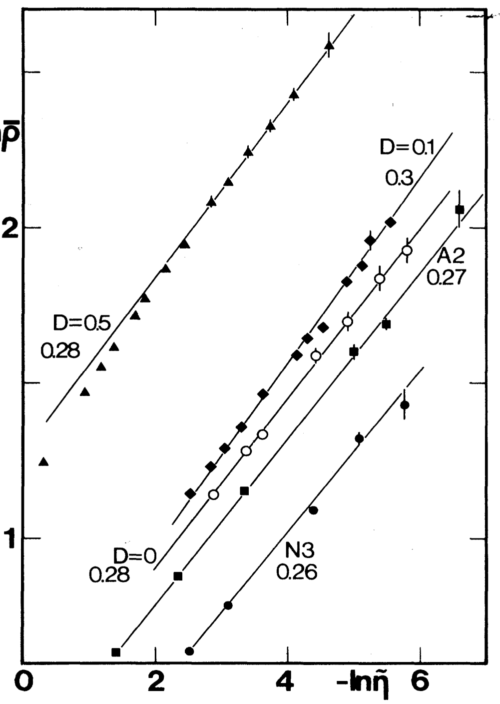

FIG. 1. Scaling plot for the steady-state density $\bar{\rho}$ vs reduced creation rate $\tilde{\eta}=(\eta-\eta_c)/\eta_c$. The lines are least-squares fits and are labeled to indicate the model and the slope; $D3$ model plots are identified by D values. For $D=0.5$, only the last six data points have been fit.

the process at site i, if it is permitted by the kinetic rules. Periodic lattices of 10 000 and 20 000 sites were employed, with run times of up to 10^5 trials per site, close to the critical point. The results for $\bar{\rho}$ are plotted versus the scaled creation rate $\tilde{\eta}=(\eta-\eta_c)/\eta_c$ in Fig. 1. A least-squares analysis yields $\beta=0.27\pm0.02$ for the $A2$ model in one dimension, consistent with RFT behavior. The kinetic rules, series expansion, and simulation results for the CP, A, $N3$, and $A2$ models are summarized in Table I. There is strong evidence for universality of critical behavior among these models. Together with earlier findings[7,12] these results indicate that the critical behavior is unaffected by a wide range of modifications of the kinetic rules.

III. COMPETING DIFFUSION AND ANNIHILATION

Given the results described in the previous section, it seems reasonable to expect RFT critical behavior in a wide variety of systems having both a scalar order parameter and a continuous transition to an absorbing state. It will be noted that the models considered above have no explicit diffusion process. However, field-theoretic analyses[8,13] and simulations[7,12] of *diffusive* Schlögl models also yield RFT behavior. There is no reason to expect that incorporation of diffusion would alter the critical exponents of the models studied in Sec. II. In fact, it is readily seen that a coarse-grained description of the nondiffusive lattice models will include a diffusive term, i.e., $\propto \nabla^2\rho$. Even without coarse graining, an effective hopping process is present in the CP and its variants, e.g., through the sequence: ●○→●●→○● (● and ○ denoting occupied and vacant sites, respectively). Thus the models are inherently diffusive, even if the bare hopping rate is zero.

Instead of devising further complications of the kinetic rules, it seems more profitable to consider a new aspect of reaction-diffusion models: competition between diffusion and multiparticle annihilation. Such competition is realized in the *triplet annihilation* or $D3$ model, which includes the processes of diffusion, autocatalytic creation, and cluster annihilation. A fraction D of the attempted moves is nearest-neighbor hopping, in which a site i is chosen at random and σ_i and σ_{i+1} are interchanged. Creation proceeds as in the CP [at rate $(1-D)\eta/(1+\eta)$ rather than η], and there is mutual annihilation in clusters of three at rate $(1-D)/(1+\eta)$. Three consecutive sites must be occupied for annihilation to occur; they are vacated simultaneously. Evidently, diffusion inhibits annihilation by dispersing clusters.

Simulations of the $D3$ model (in one dimension) were used to determine the steady-state occupation fraction, following the procedure outlined above. Lattices of 10 000 and 20 000 sites were again employed. Simulations at $D=0$ reveal a continuous transition at $\eta_c\cong6.72$, with $\beta\cong0.28$ (see Fig. 1). As D is increased from zero, the critical point shifts to smaller values of η, as expected, since diffusion tends to suppress annihilation and enhance creation by breaking up clusters. The occupation fraction $\bar{\rho}(\eta)$ is shown for several values of D in Fig. 2. Remarkably, above a critical diffusion rate, $D^*\cong0.58$, η_c is *zero*, so that an active steady state is possible for any creation rate. For $D<D^*$, an active steady state is possible for $\eta>\eta_+$, and again for $\eta<\eta_-$; for $\eta_-<\eta<\eta_+$ only $\bar{\rho}=0$ is possible. The critical line consists of two branches, η_+ and η_-, which meet at (η^*,D^*), as shown in Fig. 3. Such a reentrant phase diagram has not been found previously in reaction-diffusion systems.

It should be emphasized that the small $-\eta$, $D<D^*$ phase is not merely an artifact of slow relaxation. States in this regime have been maintained for runs of 10^6 trials per site (i.e., 10–20 apparent relaxation times) and exhibit reproducible properties which vary smoothly with η and D. It appears likely that the critical line η_- extends all the way to $D=0$; for $\eta=0.0001$, $D_c\cong0.08$, and an active steady state is presumably viable at even smaller D values for smaller creation rates. Typical configurations observed in the neighborhood of η_+ are quite different from those for $\eta=0$. In the former case, particles are strongly clustered in dense colonies, separated by expanses of empty sites; in the latter case, particles are more uniformly distributed, and colonies are sparse. The contrast illustrates alternative survival strategies for particles which catalyze their own destruction. When $\eta>\eta_+$, creation simply outpaces annihilation; for $\eta<\eta_-$, the particles evade destruction by dispersing soon after they appear. The latter strategy is viable so long as diffusion is fast, relative to creation. Viewed in the context of population dynamics, the approach to η_- is paradoxical: The "species" can maintain a *larger* steady-state population (and reduce the likelihood of extinction) by *restraining* its reproduction rate.

The critical lines in the η-D plane are η_+, η_-, and $\eta=0$. Simulation results for the large-η phase yield β values of about 0.28 (see Fig. 1), indicating that η_+ is a line of RFT-type critical points. For $D=0.5$ there is evidence of crossover from a higher apparent β, toward the RFT value, as the transition is approached. As $\eta\to 0$ one observes the scaling behavior $\bar{\rho}\propto\eta^{1/2}$, consistent with mean-field theory (see below), as shown in the inset of Fig. 2(b). Finally, the critical behavior for $\eta\to\eta_-$ is as yet unclear. The presently available data (limited to $D=0.55$), indicate that the transition is continuous, and yield $\beta=0.5\pm0.2$. Thus the $\eta=0$ critical line is of a mean-field character, while η_- appears to have nontrivial exponents, which may not be of the RFT class. More detailed studies of critical behavior are in progress.

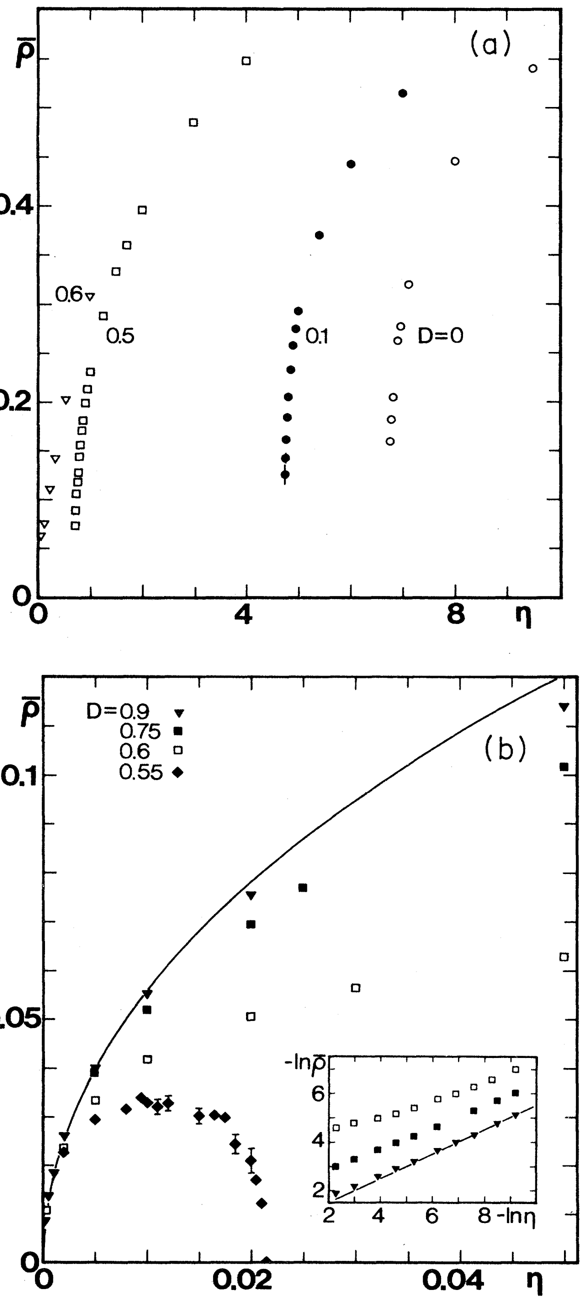

FIG. 2. Steady-state density $\bar{\rho}$ vs η in the triplet annihilation ($D3$) model for several values of the diffusion rate D. (b) Detail of the small-η regime. The inset illustrates scaling of the density as $\eta\to 0$; the slope of the straight line is $\frac{1}{2}$. Points for $D=0.75$ and 0.6 are shifted upward by 1 and 2 units, respectively, for clarity.

The phase behavior of the $D3$ model is outside the usual field-theoretic RG description of nonequilibrium critical behavior, in which the diffusion rate merely sets the scale for the temperaturelike variable.[12,13] A qualitatively correct phase diagram is, however, predicted by MFT at the pair level.[4,17] The equations for the site and nearest-neighbor pair occupation fractions (ρ and z, respectively) are

$$\dot{\rho}=\bar{\eta}(\rho-z)-\frac{3z^2(1-\bar{\eta})}{\rho} \tag{1}$$

and

$$\dot{z}=\frac{\bar{\eta}(\rho-z)(1-z)}{1-\rho}-\frac{2(1-\bar{\eta})z^2(\rho+z)}{\rho^2}+\frac{2\bar{D}(\rho-z)(\rho^2-z)}{\rho(1-\rho)}, \tag{2}$$

where a factor $(1-D)$ has been absorbed into a rescaled time variable, $\bar{\eta}=\eta/(1+\eta)$, and $\bar{D}=D/(1-D)$. The active steady-state solution is

$$\bar{\rho}=\frac{\bar{\eta}(2-\chi)+6\bar{D}}{\bar{\eta}(2\chi-1)+6\chi\bar{D}} \tag{3}$$

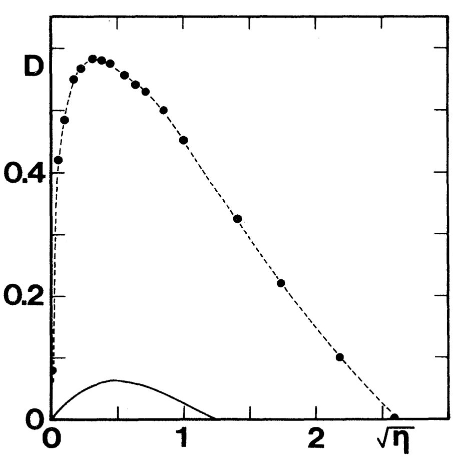

FIG. 3. Phase diagram for the triplet annihilation ($D3$) model. Points mark limiting parameter values (from simulations) below which there is no active steady state; the broken lines serve to guide the eye. The solid line is the phase boundary as predicted by pair MFT.

and $\bar{z}=\bar{\rho}/\chi$, where

$$\chi=\tfrac{1}{2}[1+\sqrt{1+(12/\eta)}]\ .$$

(This solution is locally stable.) The density scales as $\bar{\rho}\propto\eta^{1/2}$ for $\eta\rightarrow 0$ and vanishes linearly as η approaches the critical values

$$\eta_{\pm}=3[1-6\bar{D}\pm(1-12\bar{D}-44\bar{D}^{2})^{1/2}]/10 \qquad (4)$$

[the rhs of Eq. (3) is negative when $\eta_{-}<\eta<\eta_{+}$]. For $D>D^{*}\cong 0.06272$, the active steady state persists for all η. In the limit $D\rightarrow 1$, the steady-state solution takes the simple form $\bar{\rho}=\chi^{-1}$; this is in rather good agreement with the simulation, for $\eta\lesssim 0.05$ [see Fig. 2(b)]. As might be expected, MFT fails quantitatively (poor prediction for the critical line, incorrect β) away from $\eta\cong 0$.

The agreement between MFT and simulation as $\eta\rightarrow 0$ reflects suppression of correlations as the diffusion rate becomes large relative to other reaction rates. Indeed, for an Ising model with competing dynamics (spin flips at finite temperature, spin exchange or diffusion at infinite temperature) the validity of the (mean-field-like) reaction-diffusion description has been established rigorously, in the limit of an infinite diffusion rate.[18] As in the $D3$ model, the phase diagram of the Ising model with competing dynamics exhibits a dramatic change at a *finite* diffusion rate; in this case; the ferromagnetic transition (in zero field) becomes *first order* above a critical value of D.[17,19]

It is natural to ask if the three-particle annihilation rule is essential for the behavior observed in the triplet annihilation model, or whether simpler systems (e.g., diffusive versions of the CP or the binary annihilation $A2$ model) could exhibit a similar phase diagram. One should not expect the phase diagram of the contact process to change qualitatively under diffusion, since annihilation is not cooperative in this model. This conclusion is supported by pair MFT, which predicts (for a one-dimensional CP with hopping constituting a fraction D of all moves) that η_c decreases smoothly from 2 to 1 as D increases from 0 to 1. The diffusive $A2$ model presents a more delicate question, since diffusion (in one and two dimensions) is effective in creating as well as destroying pairs. Pair MFT predicts $\eta_c=2(1-3D)/(1+3D)$ for $D<\frac{1}{3}$, and $\eta_c=0$ for $D>\frac{1}{3}$, for this model in one dimension. Thus the critical line may terminate (at $\eta=0$) for sufficiently rapid diffusion, but we should not expect a distinct low-η phase.

Finally, it is of interest to examine the effect of diffusion on Schlögl's second model. A simple one-dimensional lattice version of the model (the $S2$ model) consists of the processes (1) nearest-neighbor hopping at rate D; (2) spontaneous (single-particle) annihilation at rate $(1-D)/(1+\eta)$; and (3) creation by a *pair* of particles at neighboring sites, at rate $(1-D)\eta/(1+\eta)$, of a new particle, which appears at either of the sites adjacent to the pair (if this site is vacant). The essential difference between this model and the ones studied thus far is that *two* adjacent particles are required to produce a third. So in this case diffusion competes with *creation* rather than annihilation. In the simplest (site) mean-field approximation, the steady-state density jumps from zero for $\eta<4$ to $(1+\sqrt{1-4/\eta})/2$ for $\eta>4$. However, mean-field theory at the *pair* level predicts (for $D=0$) a *continuous* transition at $\eta_c=4$. The key point is that in this approximation, as $\eta\rightarrow\eta_c$, the pair fraction $\bar{z}\propto\bar{\rho}$, not $\bar{\rho}^{2}$ as assumed in the site approximation. (Recall that simulations[12] of Schlögl's second model also reveal a continuous transition). Now for sufficiently rapid diffusion we expect $\bar{z}\propto\bar{\rho}^{2}$, i.e., diffusion can induce a discontinuous transition, as in the Ising model with competing dynamics. Such in fact is the prediction of pair mean-field theory: The transition becomes discontinuous when $D>0.198$. A test of this prediction via simulations of the $S2$ model is planned for the near future.

IV. SUMMARY

The series expansion and simulation results reported above indicate a high degree of universality in nonequilibrium critical behavior. The scope of the present work is limited in several respects: All of the models are one-dimensional, and the conclusions are based on results for the static order-parameter exponent β. More definitive conclusions on universality will require studies of higher-dimensional models, and other aspects of both static and time-dependent critical behavior. Nevertheless, the results of the present work and of earlier studies[7,8,12,13] argue strongly in support of Grassberger's conjecture that RFT is generic for single-component systems with a unique absorbing state.

In this work, the effects of competition between diffusion and multiparticle annihilation are explored for the first time, revealing a new kind of nonequilibrium phase diagram. For a sufficiently large diffusion rate $(D>D^{*})$ an active steady state is viable at any creation rate. A further surprise is the appearance, for $0<D<D^{*}$, of a distinct active steady state at very low creation rates. This phase is bounded (at $\eta=0$) by a mean-field critical line, and by a critical line (η_{-}) whose nature has yet to be determined. The phase diagrams of several other diffusive models are also discussed, in the context of mean-field theory. In particular, it is argued that for sufficiently high diffusion rates the transition in Schlögl's second model becomes discontinous. The effect of rapid diffusion on nonequilibrium phase behavior promises to be an exciting area of future investigation.

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