11-4-1996

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Kevin E. Bassler
Louisiana State University

Dana A. Browne
Louisiana State University

Recommended Citation

Bassler, K., & Browne, D. (1996). Nonequilibrium Critical Dynamics of a Three Species Monomer-Monomer Model. *Physical Review Letters, 77* (19), 4094-4097. https://doi.org/10.1103/PhysRevLett.77.4094

Nonequilibrium critical dynamics of a three species monomer-monomer model

Kevin E. Bassler and Dana A. Browne

Department of Physics and Astronomy, Louisiana State University, Baton Rouge, LA 70803

(June 11, 1996; revised August 28, 1996)

We study a three species monomer-monomer catalytic surface reaction model with a reactive steady state bordered by three equivalent unreactive phases where the surface is saturated with one species. The transition from the reactive to a saturated phase shows directed percolation critical behavior. Each pair of these reactive-saturated phase boundaries join at a bicritical point where the universal behavior is in the even branching annihilating random walk class. We find the crossover exponent from bicritical to critical behavior and a new exponent associated with the bicritical interface dynamics.

05.70.Ln, 82.20.Mj, 82.65.Jv, 64.50.Ht

Nonequilibrium models with many degrees of freedom whose dynamics violate detailed balance arise in studies of biological populations, heterogeneous catalysis, fluid turbulence, and elsewhere. The macroscopic behavior of these models can be much richer than that of systems in thermal equilibrium, showing organized macroscopic spatial and temporal structures like pulses or waves, and even spatiotemporal chaos. Even the steady state behavior can be far more complicated, involving for example generic scale invariance. Like their equilibrium cousins, nonequilibrium systems at continuous transitions between steady states show universal behavior that is insensitive to microscopic details and depends only on properties such as symmetries and conservation laws.

One of the most common continuous phase transitions in nonequilibrium models is a transition to an absorbing, noiseless steady state [1], the term absorbing indicating the state cannot be left once it is reached. Examples of this include directed percolation (DP) [2,3], the contact process [4], the dimer poisoning transition in the ZGB model [5] for the catalytic oxidation of CO, auto-catalytic reaction models [6], and branching annihilating random walks with odd numbers of offspring [7,8]. Both renormalization group calculations [2,9] and Monte Carlo simulations [3–8] show that these models form a single universality class for a purely nonequilibrium model with no internal symmetry in the order parameter.

Recently, a number of models with continuous absorbing transitions in a universality class distinct from directed percolation have been studied. These models include the probabilistic cellular automata models studied of Grassberger *et al.* [10], certain kinetic Ising models [11], the interacting monomer-dimer model [12,13], and branching annihilating random walks with an even number of offspring (BAWe) [7,14]. All of these models except for the BAWe have two equivalent absorbing states indicating the importance of symmetry of the adsorbing state to the universality class. However, the universal behavior of this new class is apparently controlled by a

dynamical conservation law. If [10] the important dynamical variables in this class are defects represented by the walkers in the BAWe model and the walls between different saturated domains in the other models, the models have "defect parity" conservation law where the number of defects is conserved modulo 2. Recent field theoretic work confirms this viewpoint [15].

We study here a simple catalytic surface reaction model with three different equivalent monomer species. This model could represent either a system with three different chemical species or an auto-catalytic reaction system in which one chemical species can adsorb on three different types of surface sites. This model has adsorbing transitions to both one and two equivalent noiseless states, and therefore is a good model to study the role of symmetry in adsorbing phase transitions. The model has two fundamental processes: (a) monomer adsorption onto sites of a substrate, and (b) the annihilation reaction of two dissimilar monomers adsorbed on nearest-neighbor sites of the substrate. Here we consider the model only in the adsorption controlled limit where process (b) occurs instantaneously. We present here results only for the one dimensional version of the model.

Calling the monomer species A, B and C, the parameters in the model are then the relative adsorption rates of the different monomer species p_A, p_B, and p_C, such that $p_A + p_B + p_C = 1$. Using static Monte Carlo simulations to find the steady state, and dynamical Monte Carlo studies described below, we find the ternary phase diagram for the model is shown in Fig. 1, where the horizontal axis corresponds to the relative adsorption rate of A and B monomers $p_{AB} = p_A/(p_A + p_B)$. There are noiseless phases where one monomer species saturates the chain occupying the corners of the phase diagram and a reactive steady state in the center. There are continuous phase transitions from the reactive phase to the saturated phases. The monomer densities undergo discontinuous, first-order, transitions from one saturated state to another. The points where the reactive phase and two

saturated phases meet are *bicritical* points [16] where two lines of continuous transitions meet a line of first order transitions. We have also constructed a mean field theory of the model, to be presented elsewhere [17], following the methods of Dickman [18]. One unusual feature of the mean field phase diagram is that the bicritical points lie on the edge of the phase diagram if the correlations are correct up to single sites or even nearest neighbor pairs. Only when the correlations up to triplets of adjacent sites are included does the bicritical point appear inside the phase diagram, indicating the importance of reproducing the correlations induced by large domains of a single saturated phase.
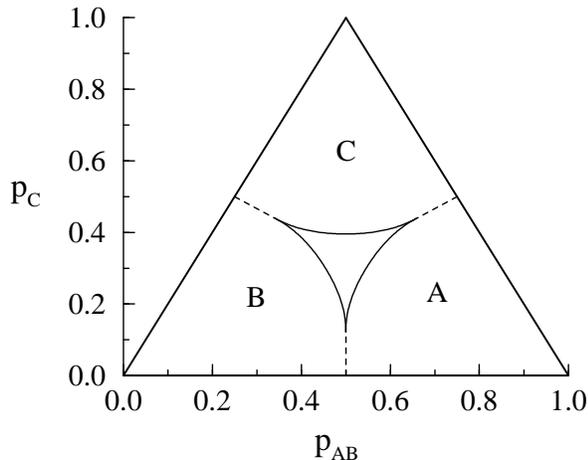

FIG. 1. Phase diagram showing three saturated phases (indicated by the letters), and a reactive phase (the unlabeled center region). Solid lines indicate continuous transitions. Dashed lines indicate first-order transitions.

We have used dynamical Monte Carlo simulations to investigate the universality classes of the continuous transitions and bicritical points, the critical dynamics of interfaces between the two symmetric saturated states at the bicritical points, the crossover behavior near the bicritical point, and the first-order lines. We use two forms of "epidemic" analysis [8,14,19] following the evolution of an initial condition chosen close to the saturated state. In the first form (defect dynamics) we use an initial condition consisting of a single vacancy in a saturated phase. The second (interface dynamics) starts from two different saturated phases separated by a single vacancy.

From the simulations we find the probability $P(t)$ the system does not fall into the saturated state in t time steps, the average number of vacancies per run $\langle n_V(t) \rangle$, and the typical size of the defect or interface per surviving run $\langle R^2(t) \rangle$. At a continuous phase transition as $t \to \infty$ it is expected that they obey power law behavior

$$P(t) \sim t^{-\delta}, \qquad \langle n_V(t) \rangle \sim t^{\eta}, \qquad \langle R^2(t) \rangle \sim t^z. \quad (1)$$

Precise estimates of the location of the critical point and

of the exponents can be made by examining the local slopes of the curves on a log-log plot and extrapolating to infinite times. Then, the effective exponent $\delta(t)$ is

$$-\delta(t) = \{\ln\left[P(t)/P(t/b)\right]/\ln b\}, \qquad (2)$$

$\eta(t)$ and $z(t)$ being defined analogously. Plotting the local slopes versus t^{-1} allows us to determine both the exponent and the critical point accurately. At the critical point the local slope will extrapolate to a constant as $t^{-1} \to 0$ with a scaling correction linear in t^{-1} [20]. Data taken away from the critical point will have local slopes that curve away from the critical point value as $t^{-1} \to 0$.
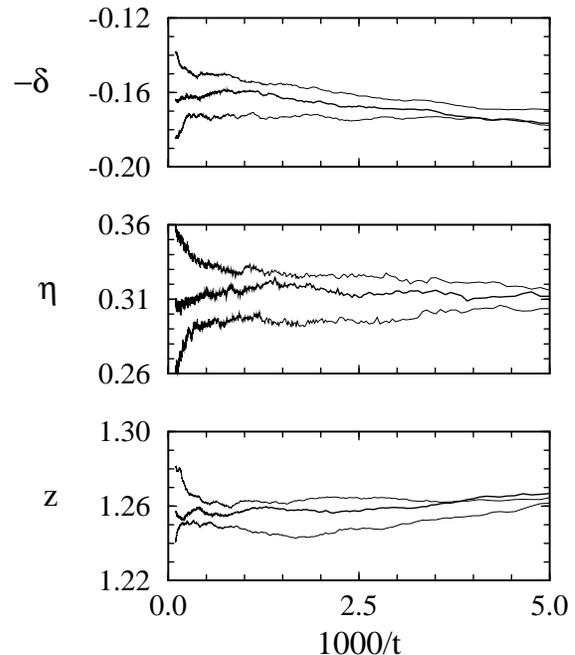

FIG. 2. Effective exponents using Eq. (2) with $b = 5$ for the defect dynamics near the critical point at $p_{AB} = 0.5$ on the line where the C poisoned phase meets the reactive phase. From top to bottom, the 3 curves in each panel correspond to $p_C = 0.3955$, 0.39575, and 0.3960, with the middle curve corresponding to the critical point.

Figure 2 shows the effective exponents of the three dynamic quantities near the phase transition to the C saturated phase at $p_{AB} = 0.5$. Using 10^5 independent runs of up to 10^4 time steps at each parameter value, we found a critical C monomer adsorption rate of $\tilde{p}_C = 0.39575(10)$, and the critical exponents are $\delta = 0.16(1)$, $\eta = 0.31(1)$, and $z = 1.255(15)$. These values are consistent with our expectation that this transition should be in the DP universality class, for which the exponents are $\delta = 0.1596(4)$, $\eta = 0.3137(10)$, and $z = 1.2660(14)$ [21]. We found similar exponents for the adsorbing transition at a number of other points along the lines separating the reactive phase and the saturated states, indicating the transition between the reactive phase and any single saturated phase
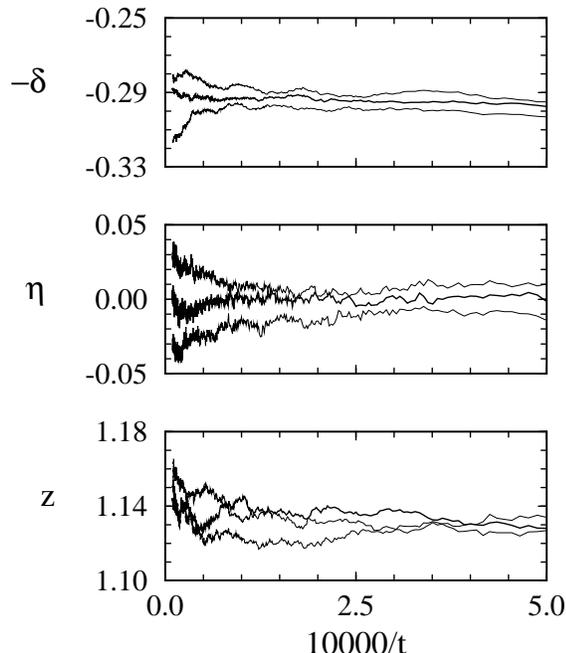
is always in the DP universality class.

FIG. 3. Effective exponents for the defect dynamics near the bicritical point where the A and B poisoned phases meet the reactive phase as defined in (2) with $b = 5$. From bottom to top, the 3 curves in each panel correspond to $p_C = 0.121$, 0.122, and 0.123, with the middle line corresponding to the bicritical point.

The same kind of analysis at the bicritical point at $p_{AB} = 0.5$, using an initial condition of a vacancy in an A-saturated phase, yields a bicritical point at $p_C = p_C^* = 0.122(1)$, and very different exponents, given the presence of two-symmetry equivalent saturated phases. From 5×10^5 runs of up to 10^5 time steps we found the local slope data shown in Fig. 3, yielding values of $\delta = 0.29(1)$, $\eta = 0.00(1)$, and $z = 1.150(15)$. These values indicate that the bicritical behavior falls in the BAWe universality class, for which $\delta = 0.285(2)$, $\eta = 0.000(1)$, and $z = 1.141(2)$ [14].

For $p_C < p_C^*$ along the A-B coexistence line, a similar analysis yields the sub-critical dynamic exponents $\delta \approx 0.5$, $\eta \approx -0.5$, and $z \approx 1$. These exponents also describe the two species version of the model which can be mapped onto the well known problem of the $T = 0$ one-dimensional kinetic Ising model for which these values are known exactly [22].

To further analyze the importance of competition in the growth of two equivalent saturated phases at the bicritical point we also studied the dynamics of an interface between those two phases. Starting with a single vacancy between the two domains, we used two different methods to analyze the behavior of the interface. Since there must always be at least one vacancy between two different sat-

urated phases, in the first method we ignore the survival probability $P(t)$ and take $\delta \equiv 0$. We then measure the number of vacancies in the interface $\langle n(t) \rangle \propto t^\eta$ and average size of the interface $\langle R^2(t) \rangle \propto t^z$. From 5×10^4 independent runs at the bicritical point, each lasting 10^5 time steps, we found the other exponents to be $\eta = 0.285(10)$ and $z = 1.14(2)$. This type of interface dynamics has been used to study the properties of critical interfaces in other models in the BAWe class, where similar results for η and z were obtained [13,14].
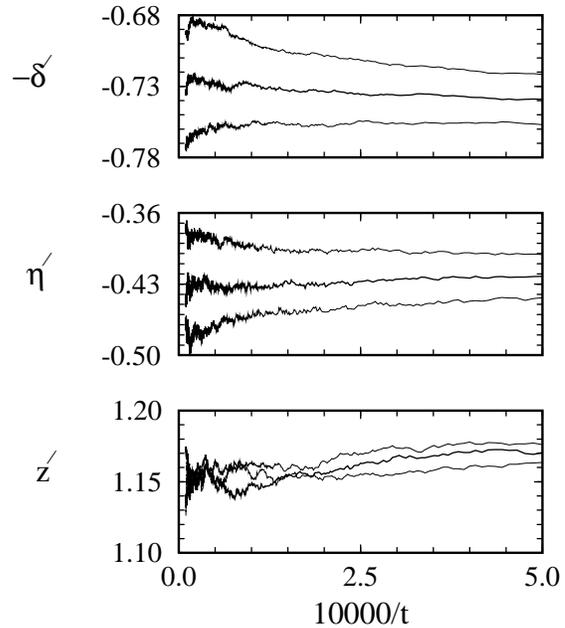
FIG. 4. Effective exponents, as in Fig. 3, for the second type of interface dynamics near the bicritical point where the A and B poisoned phases meet the reactive phase.

In the second type of interface dynamics simulations, which has not been studied before, the simulation is stopped if the interface between the domains has "collapsed" back to one vacant site. We introduce a probability of avoiding a collapse $P(t) \propto t^{-\delta'}$ and corresponding vacancy concentrations $\langle n(t) \rangle \propto t^{\eta'}$ and $\langle R^2(t) \rangle \propto t^{z'}$. Figure 4 shows results from 10^7 independent runs each lasting up to 10^5 time steps. We find values of $\delta' = 0.73(2)$, $\eta' = -0.43(2)$ and $z' = 1.15(2)$.

Note the value of the dynamic exponent z or z', which measures the size of the active region during surviving runs, is the same in both types of interface dynamics simulations as that measured for the defect dynamics. Furthermore, although the exponents δ and η are different in the three cases, their sum $\delta + \eta$ (or $\delta' + \eta'$), which governs the time evolution of the number of vacancies in just the surviving runs, seems to be the same. This indicates a universal nature of the critical spreading of the active region for models with two symmetric adsorbing states which is independent of whether defect or inter-

face dynamics is being considered. A similar result holds for some one-dimensional systems with infinitely many adsorbing states [23].

Assuming this conjecture is true, it should be noted that simulations using the first type of interface dynamics, where $\delta \equiv 0$, yield no information beyond that obtainable from simulations employing defect dynamics. However, simulations using the second type of interface dynamics measure an independent dynamic exponent δ' which we expect to be a universal number. It would be interesting to measure this exponent for other models in the BAWe class.

in the size of the interface from its smallest value is described by a new universal exponent δ'. However, the critical spreading of the reactive region, be it a defect in a single phase or a domain wall between phases, appears to be insensitive to the choice of initial conditions. This appears to result from the fact that large reactive regions are insensitive to whether the reactive regions are bounded by the same or different saturated phases.
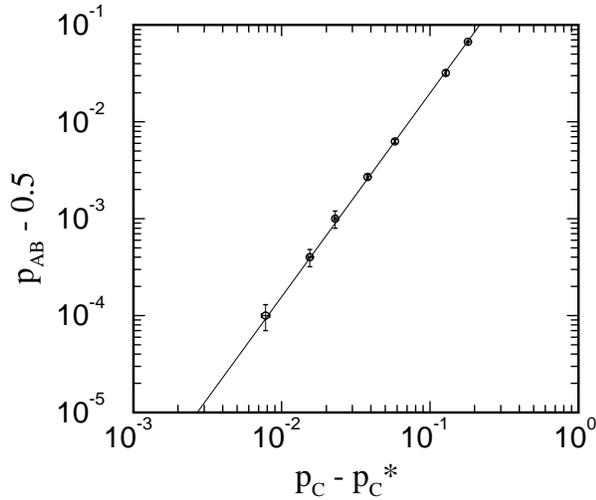
FIG. 5. Location of the critical line as a function of distance from the bicritical point. The data falls on a line with a slope corresponding to the crossover exponent $\phi = 2.1 \pm 0.1$.

Finally, we measured the crossover from bicritical to critical behavior. Near the bicritical point where the A and B poisoned phases meet, the boundary of the reactive region is expected to behave as $(p_{AB} - 0.5) \propto (p_C - p_C^*)^\phi$, where ϕ is the crossover exponent [16]. We used the dynamical simulation method to accurately determine the location of the DP phase boundary between the reactive phase and the A saturated phase near the bicritical point. From the log-log plot of $p_{AB} - 0.5$ versus $p_C - p_C^*$ shown in Fig. 5 we find $\phi = 2.1 \pm 0.1$.

We have introduced a convenient model to study the role of symmetry in the critical dynamics of adsorbing phase transitions. We have shown that the universality class of the transition changes from DP to BAWe when the symmetry of the adsorbing state is increased from one to two equivalent noiseless states. Furthermore, we have shown that having a symmetry in the adsorbing states introduces a richness into the dynamics that is not possible if there is a unique adsorbing state. In particular, the critical dynamics of the interfaces between two different adsorbing states shows a sensitivity to how the dynamics is defined, and the survival probability of fluctuations

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