

## Ziff-Gulari-Barshad model with CO desorption: An Ising-like nonequilibrium critical point

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The Ziff-Gulari-Barshad model for the reaction:  $\text{CO} + \text{O} \rightarrow \text{CO}_2$  on a catalytic surface exhibits a first-order transition to a CO-poisoned state, as the CO adsorption rate is varied. If the model is modified to include desorption of CO at rate  $k$ , the first-order transition line terminates at a critical point  $k_c$ . Here we present detailed simulations of the critical behavior. The results support theoretical arguments placing the transition in the Ising-model universality class.

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### I. INTRODUCTION

Phase transitions and critical phenomena in nonequilibrium systems have recently attracted considerable interest. Surface reaction models, in particular, have been studied intensively. One such model, devised by Ziff, Gulari, and Barshad (ZGB) [1,2], describes some kinetic aspects of the reaction  $\text{CO} + \text{O} \rightarrow \text{CO}_2$  on a catalytic surface, and exhibits two phase transitions, one continuous, the other discontinuous. The latter transition (to a CO-saturated surface) may also be continuous, if CO desorption is allowed. In this paper we examine the critical behavior associated with this transition in detail.

The transition rules for the ZGB model (a nonequilibrium lattice Markov process or *interacting particle system*) are easily stated: The surface is modeled as a regular lattice of identical adsorption sites, upon which CO and O<sub>2</sub> molecules impinge at rates  $Y$  and  $1 - Y$ , respectively. A CO molecule may adsorb at a vacant site; if it lands at an occupied site, it returns to the gas "reservoir." Oxygen molecules, by contrast, require a nearest-neighbor pair of vacant sites to adsorb. Upon adsorption O<sub>2</sub> dissociates; the two atoms may react independently. Formation of any nearest-neighbor O-CO pair (in an adsorption event) is followed immediately by a reaction to form CO<sub>2</sub> which quits the lattice, vacating two sites. (In case a newly adsorbed molecule has more than one nearest neighbor of the other species, a reacting partner is chosen at random.) Surface diffusion and nonreactive desorption were not included in the original model, but have been explored in a number of later studies.

Simulations [1,3] and mean-field theories [4–6] reveal three distinct (steady-state) phases for the two-dimensional ZGB model: For sufficiently small values of the CO adsorption rate ( $0 < Y < y_1$ ) the system eventually becomes trapped in an "oxygen-poisoned" state, i.e., every site is occupied by oxygen. At  $Y = y_1$  there is a continuous transition to an active phase (reactions proceed indefinitely), which persists up to  $Y = y_2$ , at which point there is a first-order transition to a CO-poisoned state.

O poisoning is not observed experimentally because ox-

xygen does not block CO adsorption sites. It was nevertheless of interest to establish the nature of this transition in the ZGB model. By now it has been quite clearly identified as belonging to the universality class of directed percolation [7,8]. On the other hand, numerous experiments [9–13] on catalytic surfaces reveal transitions between states of low and high CO coverage. At low temperatures there is a discontinuous drop in the rate of CO<sub>2</sub> production, as the partial pressure of CO is increased. The jump in the production rate shrinks with increasing temperature, and finally disappears at a certain critical temperature. The natural interpretation [9] is that for  $T > T_c$  the CO desorption rate is high enough to prevent occurrence of distinct high- and low-CO-coverage phases. [For the Pt(210) surface, at O<sub>2</sub> partial pressures of  $(1.3\text{--}2.0) \times 10^{-5}$  Torr, Ehsasi *et al.* [9] find  $T_c \approx 565$  K.] A number of authors [5,6,14–16] have observed that the ZGB model, modified to include CO desorption, exhibits qualitatively similar behavior. That is, if CO is permitted to desorb at rate  $k$ , then the distinction between high- and low-CO-coverage phases persists only for  $k$  less than some critical value  $k_c$ . The transition between high and low CO coverage has also been studied at the kinetic (mean-field) level using a model which is more realistic than ZGB, in that blocking of CO adsorption by O is not assumed [17]. This model again reveals a critical CO desorption rate, with the critical point appearing as a *cusp* in the bifurcation analysis.

It is of interest to study the nonequilibrium critical behavior associated with CO saturation, both in the ZGB model and in experiments on catalytic surfaces. We shall refer to this transition as "CO poisoning" even though it does not involve an absorbing state. While the ZGB model does not represent all aspects of the kinetics faithfully, the high degree of universality in critical phenomena suggests that the model can describe qualitative aspects of the critical behavior correctly. In this paper we present detailed simulation results on the critical behavior. In Sec. II we review the pertinent theoretical background at the level of mean-field theory, and then state the argument for Ising-like behavior. The simulations are described in Sec. III, and a brief summary is given in Sec. IV.

## II. BACKGROUND

To begin, we consider the simplest (one-site) mean-field description of the model [4–6,16]. In this approximation the CO and O coverages,  $\Theta_{\text{CO}}$  and  $\Theta_{\text{O}}$ , respectively, satisfy

$$\begin{aligned} \frac{d\Theta_{\text{O}}}{dt} = & 2(1-Y)(1-\Theta_{\text{O}}-\Theta_{\text{CO}})^2(1-\Theta_{\text{CO}})^3 \\ & - Y(1-\Theta_{\text{O}}-\Theta_{\text{CO}})[1-(1-\Theta_{\text{O}})^4] \end{aligned} \quad (1)$$

and

$$\begin{aligned} \frac{d\Theta_{\text{CO}}}{dt} = & Y(1-\Theta_{\text{O}}-\Theta_{\text{CO}})(1-\Theta_{\text{O}})^4 \\ & - 2(1-Y)(1-\Theta_{\text{O}}-\Theta_{\text{CO}})^2[1-(1-\Theta_{\text{CO}})^3] \\ & - k\Theta_{\text{CO}} . \end{aligned} \quad (2)$$

Here  $k$  is the CO desorption rate. These equations exhibit multiple steady-state solutions, over some range of  $Y$  values, when  $k < k_c$ . We have determined (numerically) the location of the critical point as  $k_c = 0.2170927$ ,  $Y_c = 0.6006323$ . The critical concentrations are  $\Theta_{\text{O},c} = 0.061179$  and  $\Theta_{\text{CO},c} = 0.344115$ . Let  $\Theta_{\text{O}} = \Theta_{\text{O},c} + \eta$ ,  $\Theta_{\text{CO}} = \Theta_{\text{CO},c} + \xi$ ,  $Y = Y_c + y$ , and  $k = k_c + \kappa$ . Linearizing Eqs. (1) and (2) about the critical point, we find

$$\begin{aligned} \frac{d\eta}{dt} = & -0.33229y - (1.31633 + 1.07409y)\eta \\ & - [0.49860 - 1.80723y]\xi \end{aligned} \quad (3)$$

and

$$\begin{aligned} \frac{d\xi}{dt} = & 0.96978y - 0.34412\kappa \\ & - [0.36630 + 1.57162y - \kappa]\xi \\ & - [0.96693 + 4.15833y]\eta . \end{aligned} \quad (4)$$

At the critical point ( $y = \kappa = 0$ ) the mode with eigenvalue zero is  $(\eta, \xi) = (0.354224, -0.935161)$ , and the stable mode is  $(\eta, \xi) = (0.805923, 0.592020)$ . Its decay rate is approximately 1.676. Call the amplitudes of these modes  $\phi$  and  $\psi$ , respectively. The long-time evolution of the marginally stable mode follows,

$$\begin{aligned} \frac{d\phi}{dt} = & 0.2879\kappa - 1.0155y + (1.1823y - 0.7823\kappa)\phi \\ & - 0.3703\phi^3 + \dots . \end{aligned} \quad (5)$$

This equation has the same form as for the magnetization in a mean-field theory of the Ising model near the critical point. Certain combinations of  $\kappa$  and  $y$  (i.e., the excess CO desorption and adsorption rates) serve as analogs of the temperature and external field.

At the mean-field level, the (nonequilibrium) critical point in the ZGB model with CO desorption is isomorphic to the (equilibrium) critical point in a system with a scalar order parameter. We believe that this identity of critical behavior is not simply an artifact of mean-field theory, for reasons which may be summarized as follows.

A proper coarse-grained description of the model in the neighborhood of the critical point is given by a Langevin equation for a scalar field,

$$\frac{\partial\phi(x,t)}{\partial t} = F(\phi(x',t)) + \eta(x,t) , \quad (6)$$

where  $F$  is an odd function of  $\phi$  and  $\eta(x,t)$  is a Gaussian noise with zero mean. Since the model does not have detailed balance there is no fluctuation-dissipation theorem relating the covariance of  $\eta$  to  $F$ , and we have simply  $\eta(x,t)\eta(x',t') = \delta(x-x')\delta(t-t')\Gamma(\phi(x,t))$ , where  $\Gamma$  is an even function of  $\phi$  and  $\Gamma(0) \neq 0$ . In writing Eq. (6) we assume that there is but a single mode  $\phi$  which becomes unstable at the critical point. This assumption should be valid almost everywhere in the parameter space of models exhibiting critical CO poisoning. In other words, the simultaneous vanishing of the eigenvalues associated with *two* independent modes requires “fine tuning” of reaction rules, as Grinstein, Lai, and Browne have argued in a related context [7]. The evolution of the order-parameter density is irreversible (at least on short scales), but free of any bias or driving field, and it cannot become trapped in an absorbing state. (Here one may draw a distinction between the present model and two other classes of nonequilibrium critical points with non-Ising behavior: driven diffusive systems [18,19], in which isotropy and reflection symmetry are violated, and models such as the contact process which exhibit a directed-percolation transition into an absorbing state [7].)

Grinstein, Jayaprakash, and He [20] have pointed out that reversible or not, models with short-range interactions which respect the symmetry of the lattice and symmetry under spin inversion (here,  $\phi \rightarrow -\phi$ ), belong to the Ising universality class. Critical CO poisoning should therefore be characterized by the usual Ising critical exponents. The dynamic critical behavior should be of the Ising, nonconserved order parameter (“model A” [21]) kind. We note that convincing evidence for Ising-like critical behavior in another nonequilibrium model—the majority-vote model—has recently been reported by de Oliveira [22]. Similarly, we expect Ising-like critical behavior in the “triplet creation” model with spontaneous particle creation [23]. (In the absence of spontaneous creation the latter model exhibits either a continuous or a first-order transition into an absorbing state. Spontaneous creation removes the absorbing state.)

The argument outlined above implies Ising-like critical behavior for “global” properties such as the correlation length and the order parameter. Various local properties of Ising models, such as the energy density or specific heat, have no analog in the present non-Hamiltonian model. On the other hand, it is natural to ask how the critical behavior of the reaction rate in the ZGB model is related to Ising exponents.

Let  $R$  be the rate of  $\text{CO}_2$  production per site. In a steady state  $d\Theta_{\text{CO}}/dt = 0$  implies

$$R = Y(1-\Theta_{\text{O}}-\Theta_{\text{CO}}) - k\Theta_{\text{CO}} . \quad (7)$$

Since the order parameter  $\phi$  is expected to be a linear combination of the O and CO coverages,  $R$  itself has a

linear dependence upon  $\phi$ . (It would be an extraordinary coincidence for  $R$  to have null projection on  $\phi$ .) Letting  $r = R - R_{cr}$ , we have for the singular part  $r_{sing} \propto \phi$  in the vicinity of the critical point. Now  $\phi$  is the analog of the magnetization  $m$  in the Ising model. With  $\tau = (T - T_c)/T_c$  the reduced temperature, and  $h$  the magnetic field, the critical behavior of  $m$  is given by the well-known scaling expression [24]

$$m = |\tau|^\beta M_\pm(h/|\tau|^{\beta+\gamma}), \quad (8)$$

where the branch of the scaling function  $M$  depends on the sign of  $\tau$ .  $M(x) \simeq x^{\beta/(\beta+\gamma)} = x^{1/\delta}$  as  $x \rightarrow \infty$ . Again barring a coincidence,  $\tau$  and  $h$  correspond to linear combinations of  $\kappa$  and  $y$  in the ZGB model. Thus, for example, varying  $y$  while keeping  $k$  fixed at  $k_c$  corresponds to  $h = ay$ ,  $\tau = by$ , and so to

$$r_{sing} \propto |y|^{1/\delta} \text{sgn}(y). \quad (9)$$

[There is of course one direction through the origin of the  $(\kappa, y)$  plane along which  $h = 0$ , but this direction is not known *a priori*.] We conclude that, generically, the critical behavior of the reaction rate involves the exponent  $\delta$ .

### III. SIMULATIONS

The simulation algorithm generates a sequence of trials, adsorption, and desorption, which occur with probabilities  $1-k$  and  $k$ , respectively. In the desorption process a site is selected at random; if it is occupied by CO the site is vacated, if not, the trial ends.

In the adsorption process we choose the molecule (CO or O<sub>2</sub>, with probabilities  $Y$  and  $1-Y$ , respectively), and a lattice site  $x$  random. Site  $x$  must be vacant for the trial to proceed. The CO molecule is placed at  $x$  if none of the nearest neighbors of  $x$  harbors an O atom. But if some neighbors,  $y_1, \dots, y_n$  ( $n=1, \dots, 4$ ) are occupied by O, a reacting site is selected at random from this set. The net result in this case is the emptying of one neighbor of site  $x$  while  $x$  itself remains vacant.

In the case of O adsorption, a nearest neighbor  $y$  of  $x$  is selected at random. Both  $x$  and  $y$  must be vacant for the trial to proceed. If the nearest neighbors of  $x$  are free of CO then an O atom is placed at  $x$ , but if some neighbors  $z_1, \dots, z_n$  ( $n=1, 2, 3$ ) are occupied by CO, one is selected for reaction, so that it (and site  $x$ ) are vacant at the end of the process. Adsorption and reaction at site  $y$  follow the same rules.

The model was simulated on a square lattice of  $L \times L$  sites ( $L=10-160$ ), with periodic boundary conditions. All results reported here represent steady-state conditions. A striking result of the simulations is that hysteresis is observed (for sufficiently small desorption rates  $k$ ), even in rather small systems, i.e., for  $L \geq 20$ . Examples of hysteresis in the CO coverage (for  $L=40$ ) are shown in Fig. 1. For  $k=0.05$  hysteresis is absent; this is somewhat below the critical desorption rate for this lattice size (see below). It should be noted that the coverage is stable over periods of at least  $(1-2) \times 10^4$  lattice updates, so the results represent well-defined metastable states of the model. In Fig. 2 we present CO coverage

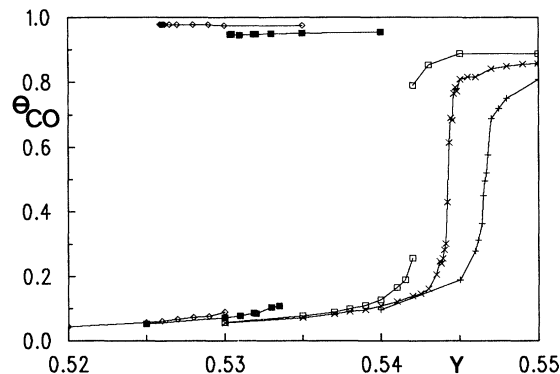


FIG. 1. Steady-state CO-coverage vs  $Y$  in simulations of the ZGB on a square lattice of size  $L=40$ . Diamonds: desorption rate  $k=0.01$ ; filled squares:  $k=0.02$ ; open squares:  $k=0.03$ ;  $\times$ :  $k=0.04$ ;  $=$ :  $k=0.05$ .

curves for larger values of  $k$ . The “susceptibility”  $d\theta_{CO}/dY$  becomes quite large in a narrow range of  $Y$  values, as one approaches  $k_c$ . A rapid decrease in the reaction rate with increasing  $Y$  (the primary experimental indication of the poisoning transition), is also seen in our simulations, as shown in Fig. 3.

Our original intent was to characterize the critical behavior of such properties as the susceptibility and the order parameter (i.e., the differences in CO or O coverage between the two phases), but it proved very difficult to obtain results of the precision required for reliable estimation of critical exponents. One source of difficulty is that unlike in simple Ising models, the coexistence curve  $Y_{coex}(k)$  is not known *a priori*. Similarly, above  $k_c$  one must determine the susceptibility over some range of  $Y$  values, to locate the maximum. In fact, a reliable determination of the value  $Y_m(k)$  which maximizes the susceptibility was feasible only for small lattices ( $L=10, 20$ ); owing to large fluctuations and slow evolution, this was

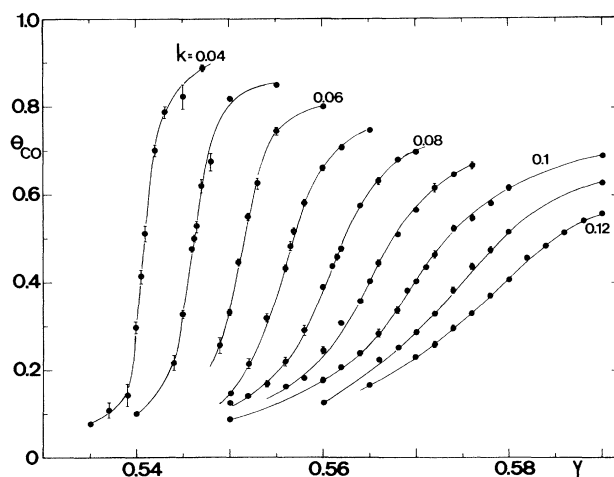


FIG. 2. CO coverage in the ZGB model,  $L=20$ , for  $k=0.04, 0.05, \dots, 0.12$ .

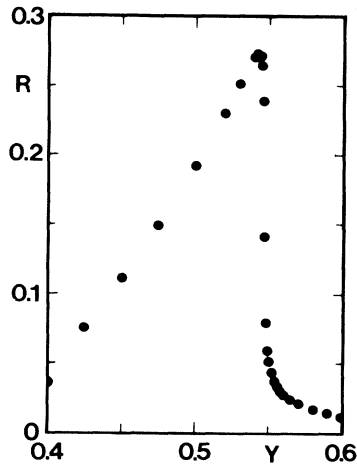


FIG. 3. CO<sub>2</sub> production rate  $R$  vs  $Y$ ,  $k=0.05$ ,  $L=40$ .

not practical for larger systems. Below the critical point, coexistence can in principle be determined using the zero interface-velocity criterion [1,5], but we found it impossible to stabilize an interface in small systems. Instead of analyzing the susceptibility or coverage gap, we turned to a coverage-histogram analysis [25] to determine the size-dependent critical desorption rate  $k_c(L)$ .

We estimate the probability distribution for  $\Theta_{\text{CO}}$  by noting the number of times  $N_i$  the coverage fell in the intervals  $[0, \Delta)$ ,  $[\Delta, 2\Delta)$ , ...,  $[1-\Delta, 1]$  ( $\Delta=0.01$ ). Plots of  $P(\Theta_{\text{CO}})$  ("histograms") are presented in Fig. 4. For sufficiently small  $k$  (depending on the lattice size), the histogram is bimodal over some narrow range of  $Y$  values, representing distinct phases. The heights of the peaks vary with  $Y$  in the expected fashion; peaks of equal height are observed at  $Y_m(k)$ , supporting the interpretation of  $Y_m(k)$  as the coexistence line. For larger  $k$  values only a

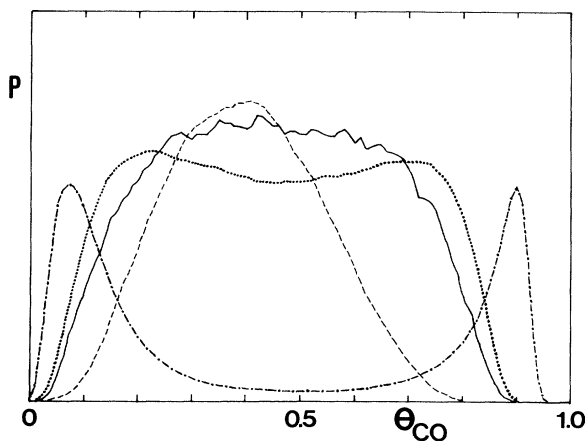


FIG. 4. Unnormalized CO coverage histograms for  $L=20$ . Broken line:  $k=0.10$ ,  $Y=0.57$ ; solid line:  $k=0.075$ ,  $Y=0.559$ ; dotted line:  $k=0.07$ ,  $Y=0.55625$ ; dashed-dotted line:  $k=0.0425$ ,  $Y=0.542$ .

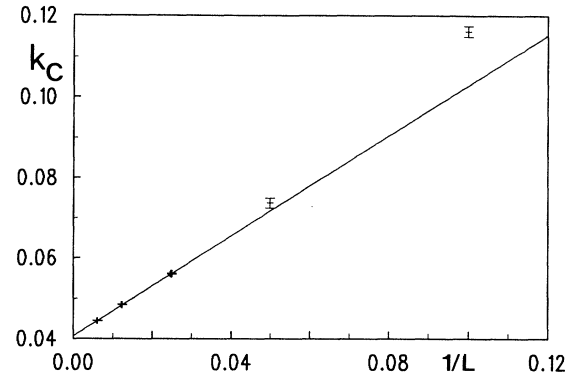


FIG. 5. Critical desorption rate  $k_c$  vs  $1/L$ .

single peak, whose location varies smoothly with  $Y$ , is observed. We locate the effective critical desorption rate  $k_c(L)$  at the boundary between these regimes. At  $k_c(L)$  (and the corresponding  $Y_m$ ), the histogram exhibits a plateau extending over a broad range of  $\Theta_{\text{CO}}$  values.

Finite-size scaling theory [26] implies that the  $L$ -dependent critical point scales with system size so

$$k_c(L) - k_c = aL^{-1/\nu}, \quad (10)$$

where  $k_c$  is the critical desorption rate in the infinite-size limit and  $\nu$  is the correlation-length critical exponent. In Fig. 5 we plot  $k_c(L)$  vs  $1/L$  for  $L=10, 20, 40$ , and  $160$ . The points (particularly those for the three largest system sizes) fall very close to a straight line, yielding the estimate  $k_c=0.04060(5)$  for  $L \rightarrow \infty$ , and confirming the exponent value  $\nu=1$ , the same as for the Ising model in two dimensions. A similar analysis yields  $Y_c=0.54212(10)$  for the critical CO adsorption rate. Our results for  $k_c$  and  $Y_c$  are consistent with those recently reported by Brosilow and Ziff [16], who performed simulations using a "constant coverage ensemble" method.

Finally, we report some preliminary results on the fourth-order cumulant of the CO coverage,

$$u_L = [\langle \Theta_{\text{CO}}^4 \rangle - 3\langle \Theta_{\text{CO}}^2 \rangle^2 - 4\langle \Theta_{\text{CO}} \rangle \langle \Theta_{\text{CO}}^3 \rangle + 12\langle \Theta_{\text{CO}} \rangle^2 - 6\langle \Theta_{\text{CO}} \rangle^4] \times \{3[\langle \Theta_{\text{CO}}^2 \rangle - \langle \Theta_{\text{CO}} \rangle^2]\}^{-1}. \quad (11)$$

For  $L=10, 20$ , and  $40$  we determined the line  $Y_u(k)$  of  $Y$  values which maximize the cumulant (at fixed  $k$ ). To within uncertainty, this line agrees with  $Y_m(k)$  defined above. For Ising models on the square lattice with periodic boundary conditions, it has been found [27] that  $u_L$  takes the value  $0.61$  at the (infinite lattice) critical temperature (and for zero external field) independent of  $L$ , so long as one is in the scaling region. The value  $0.61$  is a universal, finite-size property of Ising-like models at their critical temperature. In the present case, if the critical point is in the Ising universality class,  $u_L$  should attain this value at  $k_c$ , on the coexistence line,  $Y_u(k_c)$ . In fact

we find that  $u_L=0.61$  for  $k=0.040-0.041$ , for the lattice sizes considered. This is certainly consistent with an Ising-like critical point at  $k_c=0.0406$ , but we must regard this result as preliminary since we have been unable to obtain useful data on  $u_L$  for larger lattice sizes.

#### IV. SUMMARY

We have performed mean-field and Monte Carlo studies of the Ziff-Gulari-Barshad model incorporating desorption of CO. A general argument leads us to expect that the critical point for CO "poisoning" belongs to the Ising universality class. The critical behavior of the reaction rate is found, in general, to involve the Ising exponent  $\delta$ . Analysis of simulation results using finite-size

scaling yields a correlation-length exponent  $\nu=1$ , consistent with Ising-like critical behavior.

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