

Critical behavior of nonequilibrium continuous phase transition in $A + BC$ catalytic reaction system

Da-yin Hua

Physics Department, Ningbo University, Ningbo 315211, China

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We study two lattice gas models for the $A + BC \rightarrow AC + \frac{1}{2}B_2$ reaction system. Model I includes the influences of the adsorbate diffusion and model II includes the effect of the diffusion and position exchange of B and C atoms. Model I exhibits a continuous phase transition with infinitely many absorbing states from a reactive state to a poisoned state of B and C atoms and a discontinuous transition to a poisoned state of A and B atoms when the fraction of A in the gas phase varies. The critical exponents are estimated accurately. The simulation results indicate clearly that the critical behavior of the continuous phase transition in model I belongs to the directed percolation (DP) universality class. Model II, however, exhibits a continuous transition with two absorbing states, and its critical behavior is obviously distinct from the DP universality class.

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I. INTRODUCTION

The study of the nonequilibrium continuous phase transitions (NECPT's) in many-particle systems has attracted a great deal of interest over the past two decades, since they possess wide-ranging applications in many branches of physics, chemistry, biology, and even sociology [1,2]. Compared to their equilibrium counterpart, nonequilibrium phase transitions are much less understood due to the lack of a general framework [1,2]. One of the most remarkable progress is that the critical behaviors of the continuous phase transitions with absorbing states in many systems have been found to belong to a few universality classes. The most robust is the directed percolation (DP) class in spite of the quite dramatic differences in the microscopic processes of various models [1–14]. An important exception of the DP class is the so-called parity-conserving (PC) class [8,15–20]. On the other hand, recent work suggests that the conserved field should introduce another kind of critical behavior [21].

Recently, the influence of particle diffusion in a system on the critical behavior of the NECPT has been paid much more attention. Dickman has noted that the continuous phase transition in the contact process (CP) model including the diffusion still belongs to the DP universality class [22]. However, a study of the pair contact process with diffusion (PCPD) or annihilation-fission (AF) process $2A \rightarrow \emptyset$, $2A \rightarrow 3A$ suggests that the diffusion of the particles should introduce a new kind of critical behavior [23–32]. This model without diffusion was first investigated by Jensen, and its critical behavior of the continuous transition with infinitely many absorbing states belongs to the DP class [13].

For the basic CP model, there is only an absorbing state and the diffusion of the single particle does not change the structure of the absorbing state. However, when particle diffusion is introduced in the PCP model, the number of the absorbing states can be changed obviously and the system is trapped in only two absorbing states: one a vacuum state and the other a single-particle state.

In the present paper, we will study the influence of particle diffusion on the phase transition with absorbing states

through two modified ABC lattice models [33]. Model I includes the influences of the adsorbate diffusion but model II includes the effect of the diffusion and position exchange of B and C atoms. In our investigation by means of Monte Carlo simulations, both models exhibit a continuous phase transition from the reactive state to the poisoned state, but their critical behaviors are obviously different.

II. MODEL AND ALGORITHM

Brosilow and Ziff (BZ) have suggested an $A + BC \rightarrow AC + \frac{1}{2}B_2$ reaction net to study the NO+CO catalytic reaction system [34,35] through a simplified Langmuir-Hinshelwood (LH) mechanism. The model gives rise only to the surface species A , B , and C , which is called the ABC model [33], which was later studied by Meng, Weinberg, and Evans (MWE) [36,37].

The ABC model:

- (a) $A_g + * \rightarrow A_{ads}$,
- (b) $A_{ads} \rightarrow A_g$,
- (c) $BC_g + 2* \rightarrow B_{ads} + C_{ads}$,
- (d) $A_{ads} + C_{ads} \rightarrow AC_g + 2*$,
- (e) $B_{ads} + B_{ads} \rightarrow B_{2(g)} + 2*$.

The subscript g means a species in the gas phase, ads indicates the adsorption state of a species on the surface, and $*$ is the active site on the catalytic surface.

This model can exhibit a continuous phase transition from the reaction state into one of the infinitely many absorbing states of adsorbed C and B with the fraction y of A species varying in the gas phase. The critical behavior of the continuous transition belongs to the DP universality class when the desorption of adsorbed A is neglected [38,39]. In these investigations, the particle diffusion is neglected. Actually, the particles on the surface can diffuse rapidly in some cases and can take an important role in the dynamical behavior [40–43]. When the diffusion of adsorbed C on the surface is included, it can have an important effect on the phase diagram [44]. On the other hand, the reversible process of rapid BC dissociation can lead to a position exchange of B and C

atoms at the two nearest-neighbor (NN) sites, which may play an important role in the dynamical behavior although it is very slow.

In order to investigate the effect of the particle diffusion on the critical behavior, we study two modified models of the ABC model. In model I, only the diffusion of particle C is included cooperating with the ABC model reaction scheme. In model II, the diffusion of particles A, B, and C and the position exchange between adsorbed B and C atoms at two NN sites are considered.

We introduce two relative probabilities p_{dif} and p_{exch} of the particle diffusion and exchange processes, respectively. We carry out our simulation on a $L \times L$ triangular lattice by sequential trials of the adsorption-reaction, diffusion, and exchange processes. Initial empty lattice and periodic boundary conditions are used in our simulation.

At the beginning of a simulation step, a random number ρ_0 between 0.0 and 1.0 is generated. A diffusion attempt, exchange process, or adsorption-reaction process is performed, respectively, for $\rho_0 < p_{dif}$, $p_{dif} < \rho_0 < (p_{dif} + p_{exch})$, or $\rho_0 > (p_{dif} + p_{exch})$ following (a), (b), and (c).

(a) At the beginning of the diffusion process, a site is chosen randomly, and the trial continues if the site is occupied by a corresponding particle. Then, an adjacent site is selected randomly, and if the latter site is vacant, the particle jumps to it. If the diffusion succeeds, a reaction trial will follow as in item (c).

(b) For the exchange process, a site is chosen randomly, and the trial continues if the site is occupied by B or C. Then, an adjacent site is selected randomly, and if the latter site is occupied by another B or C, the two particles exchange their positions. If the exchange process succeeds, a reaction trial will also follow as in item (c).

(c) The adsorption-reaction process begins with a random collision of a gas molecule on a $L \times L$ triangular lattice. The colliding molecule is chosen to be a monomer (A) with a given probability y which is the fraction of A in the gas phase and a dimer (BC) with a probability $1 - y$. If a monomer A is chosen, a site is chosen randomly. If the site is occupied, the trial ends; otherwise, A adsorbs. Then the six NN sites are checked, and if there are C atoms, the adsorbed A reacts with a C atom to form AC, which desorbs at once and leaves two vacant sites. If a dimer BC is selected, a pair of NN sites is selected randomly. If either site is occupied, BC rebounds back and the trial ends. Otherwise BC dissociates and adsorbs on the two sites, and then checks the eight NN sites of the pair. The C atom reacts with an adjacent A on the NN site to form an AC which desorbs at once and leaves two vacant sites, and B reacts with another B atom on the NN site to produce a B_2 molecule which also desorbs at once and leaves two vacant sites.

III. SIMULATION RESULTS

As $p_{exch} = 0$ and only particle C can diffuse, the system evolves along the dynamic rules of model I, and we can obtain Fig. 1 as $p_{dif} = 0$. When y is near to the continuous transition point y_c , the adsorbed species on the surface are mainly C atoms and they can form domain structure easily.

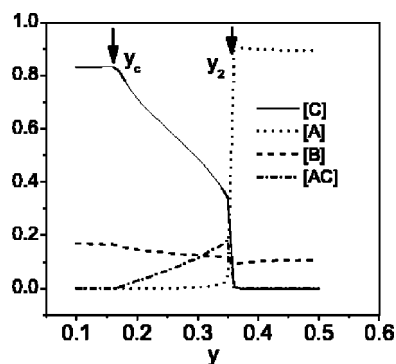


FIG. 1. The rate of AC production and the average coverage of A, C, and B are plotted as a fraction of y . Phase transitions occur at y_c and y_2 .

The diffusion of C atoms on the surface has an important influence on the domain interface, and therefore increases the opportunity of A to be adsorbed. From the simulation results in Fig. 2 for $p_{dif} = 0.1$, it shows that the critical point y_c shifts toward a lower value but the discontinuous transition point y_2 changes a little. It is obvious that the reaction window increases with the diffusion rate increasing. These simulation results are well consistent with the results of previous work [45].

It is quite difficult to directly estimate the accurate critical value y_c and corresponding critical behaviors at y_c [46] due to the critical slowing down and strong finite-size effects. In this work, we employ the finite-size scaling (FSS) method developed for the nonequilibrium continuous phase transition by Aukrust, Browne, and Webman [12,19,46] to estimate the critical point y_c , the order parameter exponent β , and other correlation length exponents.

The order parameter describing the absorbing phase transition is ρ , which is the fraction of vacant sites on the surface in the steady state; it behaves as below when y is near to the critical fraction y_c :

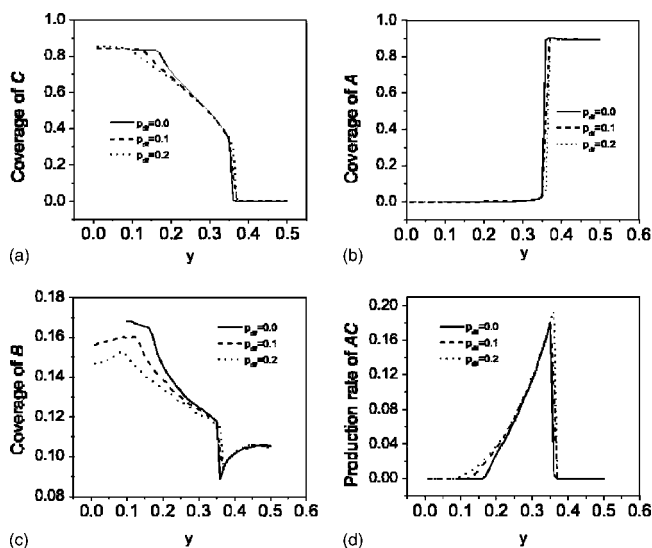


FIG. 2. The stationary coverage and production rate under different diffusion rates: (a) The coverage of C, (b) the coverage of A, (c) the coverage of B, and (d) the production rate of AC.

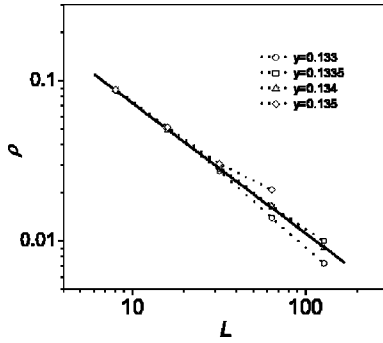


FIG. 3. The log-log plot for $\rho(y, L)$ vs L with different values of y as $p_{dif}=0.1$. The slope of the straight line that goes through the data gives an estimate of $-\beta/v_{\perp}$.

$$\rho \propto (y - y_c)^{\beta}, \quad (1)$$

where β is the order parameter exponent and the critical point y_c is accurately estimated by the FSS method. For the second-order phase transition, there are a characteristic length scale ξ and a time scale τ which denote the correlation length in space and time directions and they diverge in the neighborhood of the critical point as

$$\xi \propto |y - y_c|^{-\nu_{\perp}}, \quad (2)$$

$$\tau \propto |y - y_c|^{-\nu_{\parallel}}, \quad (3)$$

where ν_{\perp} (ν_{\parallel}) is the correlation length exponent in the space (time) direction.

At criticality, various ensemble-averaged quantities depend on the system size L through the ratio of the system size and the correlation length L/ξ . Therefore, we can take the following scaling form for the order parameter $\rho(y, L)$ in the adjacency of the critical point:

$$\rho(y, L) \propto L^{-\beta/\nu_{\perp}} f[(y - y_c)L^{1/\nu_{\perp}}], \quad (4)$$

so that, at y_c ,

$$\rho(y_c, L) \propto L^{-\beta/\nu_{\perp}} \quad (5)$$

and the scaling function

$$f(x) \propto x^{\beta} \quad (6)$$

for large x . In the supercritical region ($y > y_c$), the order parameter $\rho(y, L)$ remains finite in the limit $L \rightarrow \infty$, but it decays faster than a power law in the subcritical region ($y < y_c$).

For the characteristic time τ , we can take the following finite-size scaling form in the vicinity of y_c :

$$\tau(y, L) \propto L^z h[(y_c - y)L^{1/\nu_{\perp}}], \quad (7)$$

where $z = \nu_{\parallel}/\nu_{\perp}$ is the usual dynamical exponent. At y_c we have

$$\tau(y_c, L) \propto L^z. \quad (8)$$

We calculate the moments τ_s for each sample s which denotes a simulation entering the absorbing state from the initial empty state; therefore, we can measure the characteristic time τ following the equation

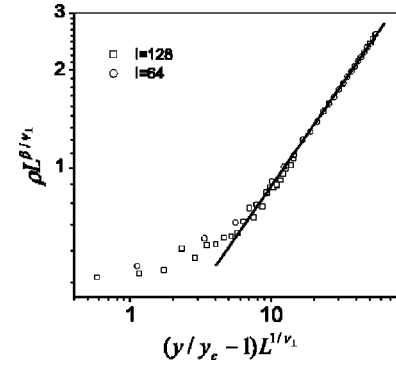


FIG. 4. The double-logarithmic plot for the data of $\rho L^{\beta/\nu_{\perp}}$ against $(y/y_c - 1)L^{1/\nu_{\perp}}$ for the two system sizes. p_{dif} is the same as that in Fig. 3.

$$\tau_s(y, L, s) = \sum_t t \rho(y, L, t, s) / \sum_t \rho(y, L, t, s), \quad (9)$$

where $\tau = \langle \tau_s \rangle_s$, and $\rho(y, L, t, s)$ is the order parameter defined above.

The system first reaches a quasi steady state, stays for a reasonably long time, and finally evolves into an absorbing state. In the simulation process, we first calculate the average of time series of the vacancy fraction over a set of surviving independent samples which have not yet entered the absorbing state when the simulation ends and then measure the stationary value of the vacancy fraction as order parameter from the average of time series. The number of Monte Carlo steps varies from 1000 as $L=8$ to 3×10^4 for $L=128$ (a Monte Carlo step refers to an attempted adsorption-reaction step on the average at every lattice site). From about 5000 independent simulations, we choose 500 surviving samples to calculate the order parameter ρ when $L=8$. The number of independent surviving samples varies from 500 for $L=8$ to 200 for $L=128$.

From Eq. (5), the data should fall on a straight line with a slope $-\beta/\nu_{\perp}$ for $y=y_c$ on a log-log plot of ρ as a function of L . In Fig. 3, we show the log-log plot of ρ as a function of L which is selected to be 8, 16, 32, 64, and 128, respectively. For our system, we find $y_c = 0.1335 \pm 0.0005$ and β/ν_{\perp}

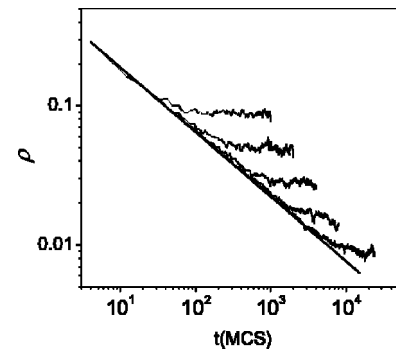


FIG. 5. The time dependence of the order parameter ρ for various sizes L at $y_c = 0.1335$. From top to bottom, the curves correspond to $L=8, 16, 32, 64, 128$. The slope of the straight line gives the value of $\beta/\nu_{\parallel} (= 0.465 \pm 0.005)$. p_{dif} is the same as that in Fig. 3.

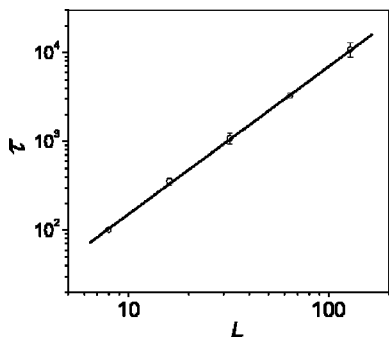


FIG. 6. The characteristic time τ against the system size L in a log-log plot. The solid line is of slope 1.68 ($=v_{\parallel}/v_{\perp}$). p_{dif} is the same as that in Fig. 3.

$=0.81 \pm 0.01$ at y_c from the slope of the straight line. This value of β/v_{\perp} is in excellent agreement with that (0.80 ± 0.01) of the DP universality class [47]. We can get further supporting results for our simulation.

In Fig. 4, we have plotted $\rho L^{\beta/v_{\perp}}$ versus $(y/y_c - 1)L^{1/v_{\perp}}$ on a log-log plot. From Eqs. (4) and (6), when x is small, the data should approach a constant, while for large x , the data should fall on a line with a slope β . It is shown that with the choices $\beta/v_{\perp} = 0.81$ and $v_{\perp} = 0.73$, the data for the two system sizes shown are well collapsed on a single curve. The solid line has a slope of 0.578 ± 0.005 , which gives the asymptotic behavior for $\rho L^{\beta/v_{\perp}}$ as $L \rightarrow \infty$.

To check our simulation results, we can also calculate the decay exponent of the order parameter at the critical point. For the time dependence of the order parameter $\rho(y_c, L, t)$ at criticality, one assumes a scaling form

$$\rho(y_c, L, t) \propto L^{-\beta/v_{\perp}} f(tL^{-v_{\parallel}/v_{\perp}}). \quad (10)$$

For $L \gg 1$ and $t \ll L^{v_{\parallel}/v_{\perp}}$, we have the relation $\rho(y_c, L, t) \propto t^{-\beta/v_{\parallel}}$. In Fig. 5, we show the double-logarithmic plot of the $\rho(t)$ as a function of time t , and then we get $\beta/v_{\parallel} = 0.465 \pm 0.005$, which is consistent with the above results. Moreover, in Fig. 6, we show the characteristic time τ as a function of L on a log-log plot. From Eq. (8), the data should fall on a line with slope $z = v_{\parallel}/v_{\perp}$ at the critical point. Every calculation result is averaged over 5000 samples, and we obtain the slope $z = 1.68 \pm 0.03$ at $y_c = 0.1335$.

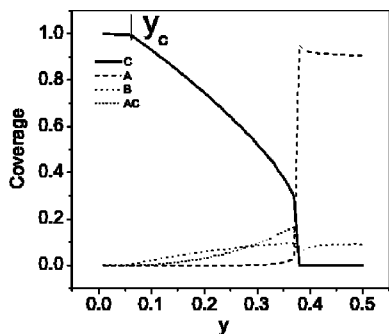


FIG. 7. The stationary coverage of A, C, and B and production rate of AC with $p_{dif} = 0.1$ and $p_{exch} = 0.2$.

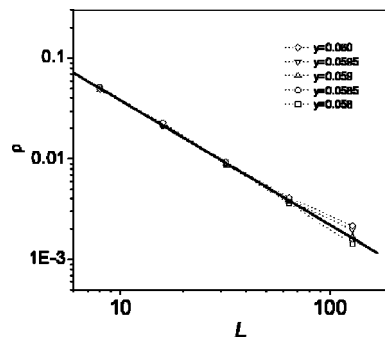


FIG. 8. The log-log plot for $\rho(y, L)$ vs L with different values of y as $p_{dif} = 0.1$, $p_{exch} = 0.2$. The slope of the straight line that goes through the data gives an estimate of $-\beta/v_{\perp}$.

Therefore, model I exhibits a continuous phase transition with infinitely many absorbing states and the critical exponents are estimated accurately, $\beta = 0.578 \pm 0.005$, $\beta/v_{\perp} = 0.81 \pm 0.01$, $\beta/v_{\parallel} = 0.465 \pm 0.005$, and $z = 1.68 \pm 0.03$. The simulation results indicate clearly that the continuous phase transition in model I belongs to the DP universality class [47] and the diffusion of particles does not change the critical behavior of the continuous phase transition. This result is expected because particle diffusion in model I does not change the structure of the absorbing states.

When the position exchange of B and C atoms on two NN sites and the influence of the diffusion of A, B, and C atoms are considered, the reaction system evolves along dynamic rule of model II. From the simulation results in Fig. 7, it is found that the system still exhibits a continuous phase transition from the reactive state to the poisoned state of C and B atoms when $p_{exch} = 0.2$. However, there is an obvious difference between Figs. 7 and 1. In Fig. 1, the coverage of C and B atoms is near to 0.83 and 0.17 in the poisoned state, respectively. But in Fig. 7, the coverage of C atom in the poisoned state is near to 1 and the coverage of B atom is near to zero. In model II, there are only two absorbing states for the continuous phase transition. One of them is that in which all sites are occupied by C atoms and another one is that in which there is only one B atom on the surface and the other sites are occupied by C atoms. It is very interesting to investigate the critical behavior of the continuous phase transition in model II.

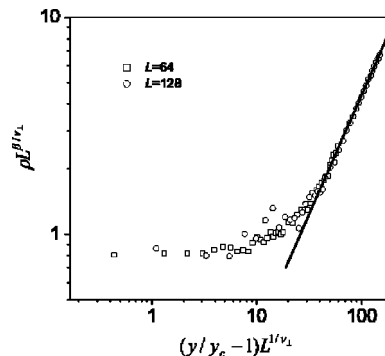


FIG. 9. The double-logarithmic plot for the data of $\rho L^{\beta/v_{\perp}}$ against $(y/y_c - 1)L^{1/v_{\perp}}$ for the two system sizes. The parameters are the same as those in Fig. 8.

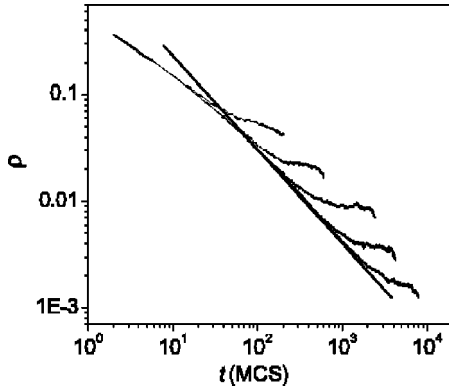


FIG. 10. The time dependence of the order parameter ρ for various sizes L at $y_c=0.059$. From top to bottom, the curves correspond to $L=8, 16, 32, 64, 128$. The slope of the straight line gives the value of β/v_{\parallel} ($=1.03\pm 0.05$). The parameters are the same as those in Fig. 8.

In Fig. 8, we show the log-log plot of ρ as a function of L which is selected to be 8, 16, 32, 64, and 128, respectively. For the system, we obtain $y_c=0.059\pm 0.0005$ and $\beta/v_{\perp}=1.28\pm 0.05$ at y_c from the slope of the straight line. This value of β/v_{\perp} is obviously distinct from that (0.80 ± 0.01) of the DP universality class [47].

In Fig. 9, we have plotted $\rho L^{\beta/v_{\perp}}$ versus $(y/y_c - 1)L^{1/v_{\perp}}$ on a log-log plot. It is shown that with the choices $\beta/v_{\perp}=1.28$ and $v_{\perp}=0.80$, the data for the two system sizes are well collapsed onto a single curve. The solid line has a slope of 1.05 ± 0.05 , which gives the asymptotic behavior for $\rho L^{\beta/v_{\perp}}$ as $L \rightarrow \infty$.

We can calculate the decay exponent of the order parameter at the critical point following the method mentioned above. In Fig. 10, we show the double-logarithmic plot of the $\rho(t)$ as a function of time t , and then we get $\beta/v_{\parallel}=1.03\pm 0.05$. Moreover, in Fig. 11, we show the characteristic time τ as a function of L on a log-log plot. Every calculation result is averaged over 5000 samples, and we obtain the slope $z=1.20\pm 0.06$ at $y_c=0.059$.

In order to understand the effect of the exchange of B and C atoms, we calculate the critical behavior when p_{exch} changes to 0.05. In Fig. 12, we show the double-logarithmic plot of the $\rho(L)$ as a function of size L . It is found that y_c

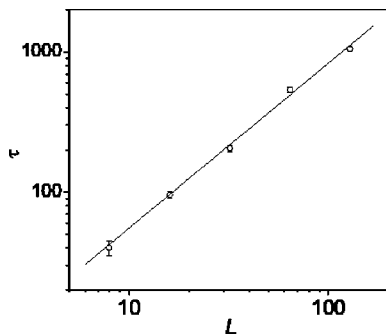


FIG. 11. The characteristic time τ against the system size L in a log-log plot. The solid line is of slope 1.20 ($=v_{\parallel}/v_{\perp}$). The parameters are the same as those in Fig. 8.

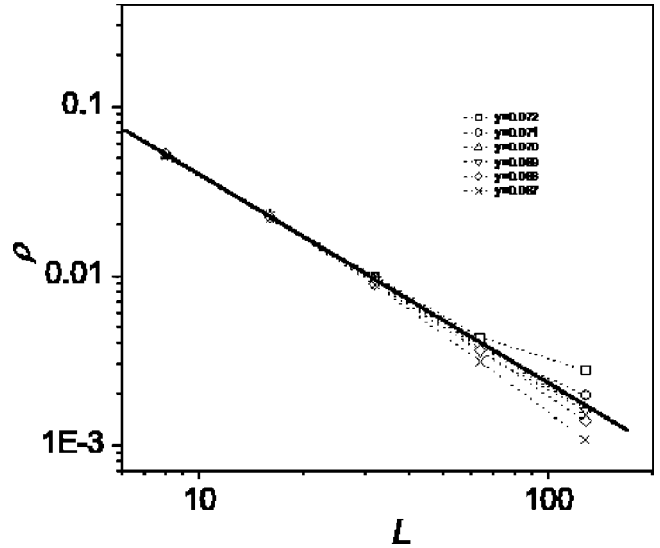


FIG. 12. The log-log plot for $\rho(y, L)$ vs L with different values of y as $p_{dif}=0.1, p_{exch}=0.05$. The slope of the straight line that goes through the data gives an estimate of $-\beta/v_{\perp}$.

$=0.070\pm 0.001$ and $\beta/v_{\perp}=1.25\pm 0.05$ at $y_c=0.070$ from the slope of the straight line. According to the same methods in Figs. 10 and 11, we can calculate $\beta/v_{\parallel}=0.94\pm 0.09$ and $z=1.24\pm 0.05$ when $y_c=0.070\pm 0.001$.

In Ref. [26], Hinrichsen has shown that the critical behavior at the transition can be affected by unusually strong corrections to scaling for the PCPD model; therefore, it becomes very difficult to estimate the critical exponents of the model accurately. In model II, there are two possible order parameters: one is that we defined above and another one is $\rho=1-\rho_c$ (ρ_c is the fraction of particle C on the surface in the steady state). Compared to Fig. 8, we show the log-log plot of ρ as a function of L for the two different order parameters in Fig. 13 at $y_c=0.059$, respectively. It is shown that, for the

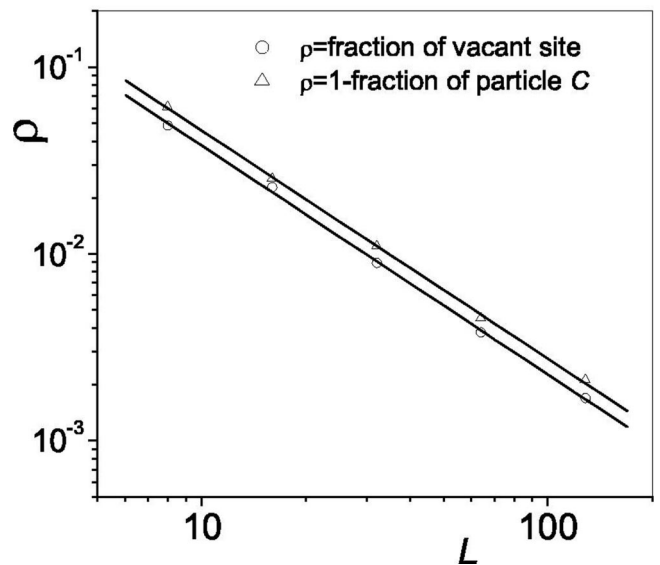


FIG. 13. The log-log plot for $\rho(y, L)$ vs L for the two different order parameters as $p_{dif}=0.1, p_{exch}=0.2$. The slope of the straight line that goes through the data gives the same estimate of $-\beta/v_{\perp}$.

two order parameters, we can obtain the same value of β/v_{\perp} accurately in the uncertainty range.

In conclusion, we studied two modified ABC models for the $A+BC \rightarrow AC + \frac{1}{2}B_2$ reaction system. Model I includes the influences of the adsorbate diffusion but model II includes the effect of the particle diffusion and the position exchange of B and C atoms. Model I exhibits a continuous transition with infinitely many absorbing states from a reactive state to a poisoned state of B and C atoms and a discontinuous transition to a poisoned state of A and B atoms with the fraction of A varying in the gas phase. The critical exponents are estimated accurately, $\beta=0.578 \pm 0.005$, $\beta/v_{\perp}=0.81 \pm 0.01$, $\beta/v_{\parallel}=0.465 \pm 0.005$, and $z=1.68 \pm 0.03$. The simulation results indicate clearly that the continuous phase transition in the model belongs to the DP universality class [47] and the diffusion of particles does not change the critical behavior of the continuous transition in model I.

Model II also exhibits a continuous transition with absorbing states. The critical exponents are estimated accurately for $p_{exch}=0.2$, $\beta=1.05 \pm 0.03$, $\beta/v_{\perp}=1.28 \pm 0.05$, $\beta/v_{\parallel}=1.03 \pm 0.05$, and $z=1.20 \pm 0.06$. We also calculate the exponent ratios $\beta/v_{\perp}=1.25 \pm 0.05$, $\beta/v_{\parallel}=0.94 \pm 0.09$, and $z=1.24 \pm 0.05$ when p_{exch} changes to 0.05. The numerical results show that there is no apparent change in the error range for the exponent ratios.

The numerical simulation shows that the critical behavior of the continuous phase transition in model II is obviously distinct from the DP universality class in model I. Furthermore, it is also different from the critical behavior of the

PCPD model studied intensively in the past several years [48,49]. Our simulation results in model II do not contradict previous investigations about the DP universality class and PCPD model [1–4,48,49]. Although the system described in model II has two absorbing states, the system only can reach one of the two absorbing states because the last adsorbed molecule must be BC before it enters into the absorbing state. Moreover, the absorbing state is not completely frozen; only the B atom can move on the surface because of the exchange of B and C atoms. Therefore, the absorbing state is not a singlet and its structure is different from the PCPD model. On the other hand, in an almost C -particle-poisoned state, with the motion of B atoms and the reaction $B_{ads} + B_{ads} \rightarrow B_{2(g)} + 2^*$ and the adsorption of BC and A , the dynamics of B particles is in analogy with a binary spreading process. However, a pair of B particles in two NN sites does not exist at the surface because of the infinite reaction rate in the model and the B atoms cannot be annihilated completely into the C poisoned state; then, its dynamics is different from the binary process in the PCPD model. A deeper investigation of the critical behavior in model II is highly desirable.

ACKNOWLEDGMENTS

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