

## Competitive reactions among three monomers over a catalytic surface

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We studied in this work a three-monomer reaction model on one- and two-dimensional lattices. We have taken different reactivity rates among pairs of monomers and the reaction between two selected monomers was forbidden. We have employed the mean field and the pair approximation to decouple the equations of motion for the densities of single and pairs of monomers. We found the stationary states and the phase diagram of the model. We have shown that, in two dimensions and within the pair approximation, there is a first-order transition line between active and poisoned steady states.

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### I. INTRODUCTION

Lattice models of catalytic reactions have attracted increasing interest since the pioneering work of Ziff, Gulari, and Barshad, known in the literature as the ZGB model [1], which mimics the oxidation of the carbon monoxide over a metal surface. The phase diagrams of a variety of models considered in the recent years, through analytical and numerical simulations, exhibit absorbing states, that is, configurations from which the system cannot escape anymore [2]. In the ZGB surface reaction model, the CO and O<sub>2</sub> molecules adsorb in a square lattice following the Langmuir-Hinshelwood mechanism. The algorithm, which describes the above reaction model, is characterized by the following steps:

- (1)  $\text{CO(g)} + V \rightarrow \text{CO(a)}$ ,
- (2)  $\text{O}_2(\text{g}) + 2V \rightarrow 2\text{O(a)}$ ,
- (3)  $\text{CO(a)} + \text{O(a)} \rightarrow \text{CO}_2(\text{g}) + 2V$ ,

where the labels g and a denote a gaseous phase and an adsorbed reactant on the surface, respectively.  $V$  indicates a vacant site. The steps (1) and (2) represent the adsorption of the reactant gases on the surface, and the step (3) the proper reaction between nearest-neighbor CO and O species. In the adsorption controlled limit, only a single parameter is necessary to describe the whole process, which is taken as the relative adsorption rate of CO molecules, denoted by  $y_{\text{CO}}$ . The results of their simulation for the square lattice, show that the system exhibits two phase transitions between active and nonactive states: for  $y_{\text{CO}} \leq y_1$ , an O poisoned state is found, and a CO poisoned state appears for  $y_{\text{CO}} \geq y_2$ . A reactive steady-state occurs for  $y_1 \leq y_{\text{CO}} \leq y_2$ , where a non-zero number of vacant sites is present in the lattice. At  $y_1$ , the densities of O and CO change continuously, while at  $y_2$  the same densities change abruptly, indicating a first-order transition. Dickman [3], employing the site and the pair approximations, within the master equation approach, also found the phase diagram for the ZGB model. In his pair approximation calculations the values determined for  $y_1$  and  $y_2$  agree very well with the corresponding simulation values.

Bassler and Browne [4] introduced a monomer-monomer reaction model with three different monomer species. They

used static and dynamic Monte Carlo simulations to determine the phase diagram of the model for the linear chain. The model presents continuous and first-order phase transitions. In their ternary phase diagram, three absorbing phases are found, and a small reactive region is present at the center of the diagram. The transitions between saturated phases are discontinuous, but the transitions between the reactive state and the absorbing states are continuous. For a description of this model only two parameters are needed, and all the reaction rates are identical for every pair of nearest neighbor pair of monomers. In a following paper [5] they also performed mean-field calculations including up to triplets of adjacent sites. With this improvement in their calculations they were able to show the appearance of a bicritical point at the end of the first-order line separating the two poisoned steady states.

In this paper we also consider a three monomer model on a lattice in what we restrict the set of all possible reactions. We take different reactivity rates among pairs of monomers, and a chosen reaction, for instance, between  $B$  and  $C$  monomers is forbidden to occur. That is, we assume that  $A$  can react with  $B$  and  $C$ , with different reactivity rates, but the reaction  $B + C$  is not permitted. This is different from the original work of Bassler and Browne [4] where the reaction between  $B$  and  $C$  is also permitted and all the reactivity rates are equal. We used the mean-field and the pair approximation calculations, in order to obtain the stationary states and the phase diagram of the model. We have shown that, in one dimension, the mean-field and the pair approximation give only a reactive phase at a single point, while in two dimensions, and within the pair approximation, at the same single point of the one-dimensional case, we found a coexistence of active and nonactive phases. In Sec. II, we introduce the model and present the mean field and pair approximation calculations for the linear chain. In Sec. III, we extend our previous analysis to the square lattice and we exhibit the phase diagram of the model. Finally, in Sec. IV, we present our conclusions.

### II. MODEL AND LINEAR CHAIN CALCULATIONS

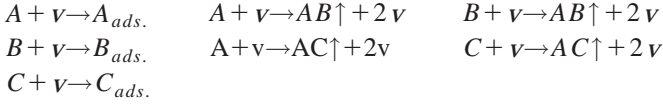
We consider a linear chain immersed in a reservoir of monomers of the type  $A$ ,  $B$ , and  $C$ . Each site of the lattice is occupied by a single monomer or it is vacant. We define the monomer densities  $p_i = N_i/N$ , where  $i$  denotes  $A$ ,  $B$ , or  $C$ .  $N_i$

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and  $N$  are the number of sites occupied by monomers of the type  $i$  and the total number of sites of the lattice, respectively. We also have the obvious constraint:

$$p_A + p_B + p_C + p_v = 1, \quad (1)$$

where  $p_v$  is the fraction of vacant sites. We also define  $y_i$  as being the probability that the next monomer arriving at the lattice is of the type  $i$ . We have  $y_A + y_B + y_C = 1$ , and we introduce the parameter  $r$  such that  $y_B = r(1 - y_A)$ . We assume that the  $B$  and  $C$  monomers cannot react themselves. So, in the table below, we list the allowed reactions that are necessary to describe the evolution of the densities in the site approximation



where, for instance,  $A + v \rightarrow AB\uparrow + 2v$ , means that the  $A$  monomer arrives at a vacant site, and finds at least one  $B$  monomer in its neighborhood. Then, they react forming the  $AB$  dimer, which leaves the lattice creating two new vacant sites.  $A_{ads}$  denotes an adsorbed  $A$  monomer in the lattice.

We now introduce a new parameter  $\beta$ , which is related to the probability of  $A$  choose  $B$  or  $C$  to react. Then, we define

$$\Pi_{AB} = \frac{\beta}{1 + \beta}, \quad (2)$$

$$\Pi_{AC} = \frac{1}{1 + \beta}, \quad (3)$$

where  $\Pi_{AB}$  is the probability of the  $A$  and  $B$  reaction, in the presence of  $C$ . In this work, we need three independent parameters to describe the stationary states of the model, that is,  $y_A$ ,  $r$  and  $\beta$ . In this way we obtain the following equations for the time evolution of the densities:

$$\frac{dp_A}{dt} = p_v \{ y_A (p_A + p_v)^2 - (y_B + y_C) [1 - (1 - p_A)^2] \}, \quad (4)$$

$$\begin{aligned} \frac{dp_B}{dt} &= p_v [y_B (1 - p_A)^2 - y_A p_B^2] - 2y_A p_v p_B \\ &\times \left( p_A + p_v + p_C \frac{\beta}{1 + \beta} \right), \end{aligned} \quad (5)$$

$$\begin{aligned} \frac{dp_C}{dt} &= p_v [y_C (1 - p_A)^2 - y_A p_C^2] - 2y_A p_v p_C \\ &\times \left( p_A + p_v + p_B \frac{1}{1 + \beta} \right). \end{aligned} \quad (6)$$

Introducing the new variables,  $\Delta = p_B + p_C$ ,  $\delta = p_B - p_C$  and  $\gamma = p_A - p_B - p_C$ , we can write the above set of equations in the following form:

$$\frac{d\Delta}{dt} = p_v \{ (1 - y_A) (1 - \gamma - \Delta)^2 - y_A [1 - (1 - \Delta)^2] \}, \quad (7)$$

$$\begin{aligned} \frac{d\delta}{dt} &= p_v (2r - 1) (1 - y_A) (1 - \gamma - \Delta)^2 - p_v \left[ \frac{y_A}{2} \left( \frac{\beta - 1}{\beta + 1} \right) \right. \\ &\left. \times (\Delta^2 - \delta^2) - y_A \delta (\Delta - 2) \right], \end{aligned} \quad (8)$$

$$\frac{d\gamma}{dt} = p_v (2y_A - 1). \quad (9)$$

It is easy to see that the steady states are given by  $p_v = 0$  or  $y_A = 1/2$ . The case  $p_v = 0$ , means that the linear chain is completely poisoned by  $A$  monomers or  $B$  and  $C$  monomers. We studied the stability of the solutions of the above system of equations by linearizing them, and considering the eigenvalues associated with the Jacobian matrix. We found that if  $y_A > 1/2$ , the lattice is poisoned by  $A$  monomers, while if  $y_A < 1/2$ , the lattice is poisoned by the  $B$  and  $C$  monomers. In the latter case, the relative amounts of  $B$  and  $C$  monomers depend on the values of  $r$ ,  $\beta$  and on the initial boundary conditions. For the particular value  $y_A = 1/2$ , we found an active state, for which the number of vacant sites is different from zero. In this case, the variable  $\gamma$  is a constant of motion, and it is given by the initial conditions. Then, we may easily write the stationary density values. For instance, the value of  $p_A$ , at  $y_A = 1/2$ , is given by

$$p_A = \frac{1}{2} (2 + \gamma - \sqrt{2 - \gamma^2}). \quad (10)$$

The  $p_B$  and  $p_C$  concentrations depend on the values of the  $r$  and  $\beta$  parameters. In particular, at the active steady state  $y_A = 1/2$ , it is easy to show that the equation

$$r = \frac{1}{2} + \frac{1}{4} \left( \frac{\beta - 1}{\beta + 1} \right) \left( \frac{2 - \gamma - \sqrt{2 - \gamma^2}}{\sqrt{2 - \gamma^2} - \gamma} \right)^2 \quad (11)$$

gives equal values for the densities of  $B$  and  $C$  monomers. We show in Fig. 1, a plot of the above equation for three different initial conditions. Below each curve, the concentration of  $C$  monomers is greater than that of  $B$  monomers, while above it happens opposite.

We turn now to the pair approximation scheme for the linear chain. In this approximation, we introduce the pair probability  $p_{ij}$  that a randomly chosen nearest neighbor pair of sites are occupied by  $i$  and  $j$  monomers or that they are vacant. In the previous treatments of the method [3,5], the rate of change of  $p_{ij}$  was evaluated by counting the changes in the number of nearest-neighbor pairs in a neighborhood of sites centered on, and including, the center pair ( $ij$ ). Here, we use a simplification of the method, where changes outside the center pair are ignored. We need to consider only the changes at a particular center pair [6]. In our case the possible pairs of nearest-neighbors are shown below.

	A	B	C	v
A	AA	×	×	Av
B	×	BB	BC	Bv
C	×	CB	CC	Cv
v	vA	vB	vC	vv

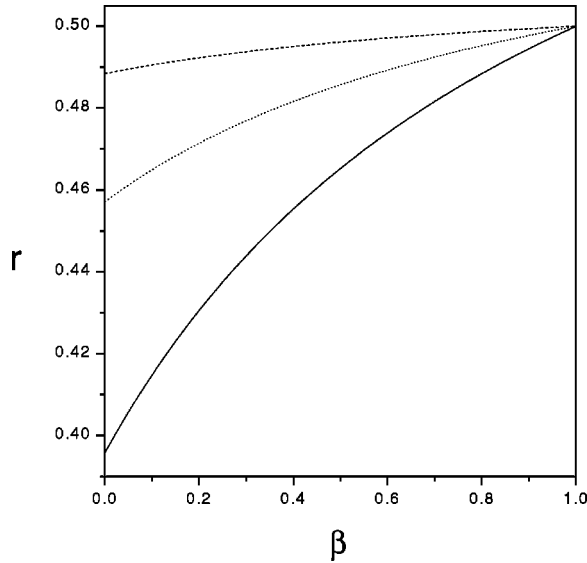


FIG. 1. Plot of the active state,  $y_A = 1/2$ , for a linear chain in the site approximation, and for three different initial boundary conditions.  $r$  is the ratio between the adsorption rates of the  $B$  and  $C$  monomers, and  $\beta$  is the probability of  $A$  choose  $B$  or  $C$  to react. Below each line the density of  $C$  monomers is greater than that of  $B$  monomers. The initial conditions are  $\gamma = -0.5$ , continuous line;  $\gamma = 0$ , dotted line;  $\gamma = 0.5$ , dashed line.

The crosses mean that the corresponding pairs are forbidden because we are working in the adsorption-controlled limit. Then, we can write the following equations relating the densities of monomers and pairs:

$$p_A = p_{AA} + p_{Av}, \quad (12)$$

$$p_B = p_{BB} + p_{BC} + p_{Bv}, \quad (13)$$

$$p_C = p_{CC} + p_{CB} + p_{Cv}, \quad (14)$$

$$p_v = p_{vv} + p_{vA} + p_{vB} + p_{vC}. \quad (15)$$

The pair densities satisfy the constraint

$$1 = p_{vv} + p_{AA} + p_{BB} + p_{CC} + 2(p_{vA} + p_{vB} + p_{vC} + p_{BC}). \quad (16)$$

In the next table, we exhibit all the possible transitions between pairs.

From \ To	vv	AA	BB	CC	vA	vB	vC	BC
vv	×	×	×	×	7	9	12	×
vA	1	4	×	×	×	×	×	×
vB	2	×	5	×	×	×	×	15
vC	3	×	×	6	×	×	×	16
AA	×	×	×	×	8	×	×	×
BB	×	×	×	×	×	10	×	×
CC	×	×	×	×	×	×	13	×
BC	×	×	×	×	×	11	14	×

We have assigned arbitrary labels for the transition rates, and again the  $\times$  mean forbidden transitions. For instance, the equation of motion for  $p_{AA}$  and for  $p_{BC}$  are written as

$$\frac{dp_{AA}}{dt} = 2T_8 - 2T_4, \quad (17)$$

$$\frac{dp_{BC}}{dt} = T_{11} + T_{14} - T_{15} - T_{16}, \quad (18)$$

where  $T_1$  to  $T_{16}$  are the transition rates. The expressions for the transition rates  $T_i$  are very simple ones in one dimension. For instance, the transition rate for the process  $BB \rightarrow vB$ , is given by

$$T_5 = y_A p_{BB} \frac{p_{vB}}{p_B} \left( \frac{p_{vv} + p_{vA}}{p_v} + \frac{p_{vB}}{2p_v} + \frac{p_{vC}}{p_v} \frac{\beta}{1 + \beta} \right). \quad (19)$$

Contrarily to the mean-field calculations, we cannot obtain analytical results in this pair approximation. We solved our set of seven nonlinear coupled equations by the fourth-order Runge-Kutta method, and we analyzed the stability of the solutions. The results we have obtained for the stationary states are very similar to those found in the site approximation. If  $y_A < 1/2$ , the linear chain is poisoned by  $B$  and  $C$  species, whose densities depend on the values of  $r$  and  $\beta$ . If  $y_A > 1/2$ , the coverage is due only to  $A$  monomers. In this pair approximation we also find an active state at  $y_A = 1/2$ . Aside small numerical differences, the plot we obtain in this approximation is quite similar to that of Fig. 1. For instance, in the active state,  $y_A = 1/2$ , the concentration of  $A$  monomers is larger than that found in the site approximation. However, as we will show in the next section, the site and the pair approximations give, at  $y_A = 1/2$ , qualitatively different results when applied to the square lattice.

### III. SQUARE LATTICE CALCULATIONS

Now the monomers  $A$ ,  $B$  and  $C$  can sit on the sites of a square lattice. The model we consider is the same one studied in the previous section. However, to take into account the different number of neighbors of  $B$  and  $C$  monomers around at an arriving  $A$  monomer, we rewrite the following relations for the probability of  $A$  choose any  $B$  or  $C$ :

$$\Pi_{AB} = \frac{\beta N_B}{N_C + \beta N_B}, \quad (20)$$

$$\Pi_{AC} = \frac{N_C}{N_C + \beta N_B}, \quad (21)$$

where  $N_B$  and  $N_C$  are the numbers of  $B$  and  $C$  monomers, respectively, which are nearest neighbors of an arriving  $A$  monomer. Again, the parameter  $\beta$  is related to the probability of  $A$  choose a selected  $B$  or  $C$  to react. Also, in two dimensions, the possible reactions and the possible pairs are the same which appeared in the one-dimensional calculations. Following the same steps, as in the linear chain calculations, we can write the following time evolution equations for the densities:

$$\frac{d\Delta}{dt} = p_v \{ (1 - y_A)(1 - \gamma - \Delta)^4 - y_A [1 - (1 - \Delta)] \}, \quad (22)$$

$$\frac{d\delta}{dt} = p_v(2r-1)(1-y_A)(1-\gamma-\Delta)^4 + p_v y_A \frac{\delta}{2} [\Delta^3 - 6\Delta^2 + \Delta(\delta^2 + 12)] - p_v y_A [\delta(\delta^2 + 4) - \psi], \quad (23)$$

$$\frac{d\gamma}{dt} = p_v(2y_A - 1), \quad (24)$$

where  $\psi$  is given by

$$\begin{aligned} \psi = & \frac{3}{8}(\Delta^2 - \delta^2)(9\Delta^2 - 16\Delta - \delta^2 + 8) \left( \frac{\beta-1}{\beta+1} \right) + \frac{3}{2}(1-\Delta) \\ & \times (\Delta + \delta)(\Delta - \delta)^2 \left( \frac{\beta-2}{\beta+2} \right) + \frac{1}{4}(\Delta + \delta)(\Delta - \delta)^3 \left( \frac{\beta-3}{\beta+3} \right) \\ & + \frac{3}{2}(1-\Delta)(\Delta - \delta)(\Delta + \delta)^2 \left( \frac{2\beta-1}{2\beta+1} \right) + \frac{1}{4}(\Delta - \delta) \\ & \times (\Delta + \delta)^3 \left( \frac{3\beta-1}{3\beta+1} \right). \end{aligned} \quad (25)$$

The stationary states are given by  $p_v=0$ , and the stability conditions assert that, if  $y_A < 1/2$ , the lattice will be covered by  $B$  and  $C$  species, while, if  $y_A > 1/2$ , the lattice will be poisoned by only  $A$  monomers. For the particular case  $y_A = 1/2$  we get an active stationary state. If we put  $\delta=0$ , which means equality of the  $B$  and  $C$  densities at the active state, then we arrive at an equation  $r(\beta)$ , which is drawn, for three different initial conditions, in Fig. 2. Each line in this figure separates the regions which are richer in the  $B$  or  $C$  species. Below each curve, the concentration of  $C$  monomers is greater than the concentration of  $B$  monomers.

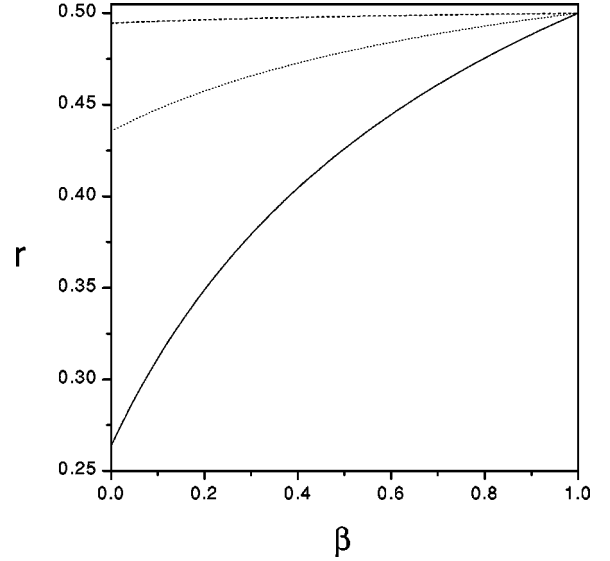


FIG. 2. Same legend as Fig. 1, except that the deposition of monomers is over a square lattice.

Finally, we briefly describe the pair approximation for the square lattice. Following the same reasoning of the previous section, we arrive at the equations of motion for the pair densities, as a function of the transition rates  $T_i$ . However, in this case, the  $T_i$  expressions are very lengthy. For instance, the expression for the  $T_5$  transition rate is

$$T_5 = y_A p_{BB} \frac{p_{vB}}{p_B} (2f + g), \quad (26)$$

where

$$\begin{aligned} f = & \frac{p_{vB}}{p_B} \left( \frac{p_{vV} + p_{vA}}{p_v} \right)^2 + \frac{p_{BB}}{2p_B} \left( \frac{p_{vV} + p_{vA}}{p_v} \right)^2 + \frac{2\beta}{1+2\beta} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) \left( \frac{p_{BB}}{p_B} \frac{p_{vC}}{p_v} + \frac{p_{BC}}{p_B} \frac{p_{vB}}{p_v} \right) + \frac{p_{vB}^2}{p_v p_B} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) \\ & + \frac{2\beta}{1+2\beta} \frac{p_{vB}}{p_B} \frac{p_{vB}}{p_v} \frac{p_{vC}}{p_v} + \frac{\beta}{1+\beta} \frac{p_{vC}}{p_v} \left( \frac{p_{BB}}{2p_B} \frac{p_{vC}}{p_v} + \frac{p_{vB}}{p_v} \frac{p_{BC}}{p_B} \right) + \frac{p_{vB}^2}{3p_v^2} \frac{p_{vB}}{p_B} + \frac{\beta}{1+3\beta} \frac{p_{vB}}{p_v} \left( \frac{p_{vB}}{p_v} \frac{p_{BC}}{p_B} + 2 \frac{p_{BB}}{p_B} \frac{p_{vC}}{p_v} \right) \\ & + \frac{2p_{BB}}{3p_B} \frac{p_{vB}}{p_v} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) + \frac{p_{vB}^2}{4p_v^2} \frac{p_{BB}}{p_B} + \frac{\beta}{1+\beta} \frac{p_{BC}}{p_B} \left( \frac{p_{vV} + p_{vA}}{p_v} \right)^2 + \frac{\beta}{3+\beta} \frac{p_{vC}^2}{p_v^2} \frac{p_{BC}}{p_B} + \frac{2\beta}{1+\beta} \frac{p_{vB}}{p_B} \frac{p_{vC}}{p_v} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) \\ & + \frac{\beta}{2+\beta} \frac{p_{vC}}{p_v} \left[ \frac{p_{vB}}{p_B} \frac{p_{vC}}{p_v} + 2 \frac{p_{BC}}{p_B} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) \right] \end{aligned} \quad (27)$$

and

$$\begin{aligned} g = & \left( \frac{p_{vV} + p_{vA}}{p_v} \right)^3 + \frac{3\beta}{1+\beta} \frac{p_{vC}}{p_v} \left( \frac{p_{vV} + p_{vA}}{p_v} \right)^2 + \frac{3p_{vB}}{p_v} \left[ \frac{2\beta}{1+2\beta} \frac{p_{vC}}{p_v} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) \right] + \frac{3p_{vB}^2}{p_v^2} \left[ \frac{1}{3} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) + \frac{\beta}{1+3\beta} \frac{p_{vC}}{p_v} \right] + \frac{p_{vB}^3}{4p_v^3} \\ & + \frac{3\beta}{2+\beta} \frac{p_{vC}^2}{p_v^2} \left( \frac{p_{vV} + p_{vA}}{p_v} \right) + \frac{\beta}{3+\beta} \frac{p_{vC}^3}{p_v^3} + \frac{3\beta}{2+2\beta} \frac{p_{vB}}{p_v} \frac{p_{vC}^2}{p_v^2} + \frac{3p_{vB}}{2p_v} \left( \frac{p_{vV} + p_{vA}}{p_v} \right)^2. \end{aligned} \quad (28)$$

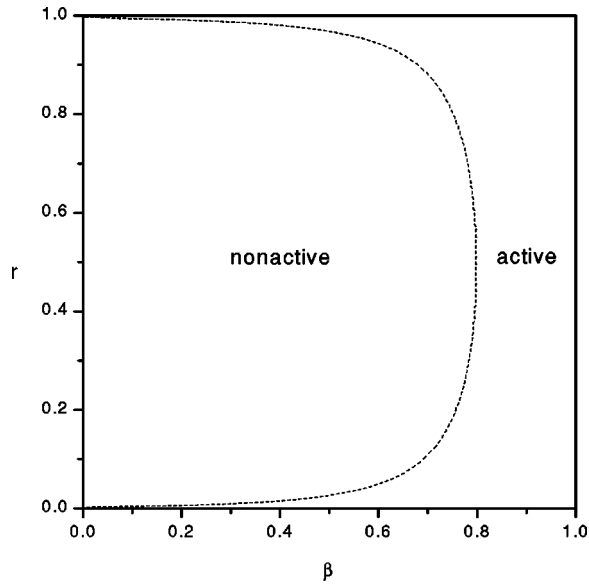


FIG. 3. Phase diagram for the square lattice, at  $y_A = 1/2$ , showing active and nonactive steady states in the pair approximation. The line between the steady states is a first-order transition line.

Again, we solved the system of seven non-linear rate equations by the fourth-order Runge-Kutta method, and we studied the stability of the possible solutions. We also found that, for  $y_A < 1/2$ , the steady states are poisoned states of  $B$  and  $C$  monomers, while for  $y_A > 1/2$ , the square lattice is poisoned by  $A$  monomers. The interesting situation arises for  $y_A = 1/2$ . The site approximation gives only active states, while in the pair approximation, we found a phase diagram in the plane  $r$  versus  $\beta$  exhibiting active and nonactive states, separated by a first-order transition line. This is indicated in Fig. 3 by the dashed line. This result is completely different from the one we have obtained in our one-dimensional calculation. At the special point  $y_A = 1/2$ , in one dimension, only an active phase was found.

#### IV. CONCLUSIONS

We have considered a three monomer reaction model on a linear chain and on a square lattice. The reaction rates among monomers were arbitrarily taken, and a selected reaction ( $B + C$ ) was forbidden to occur. We determined the rate equations for the densities of monomers and vacant sites, as well as, for the densities of all nearest neighbor pairs, through the master equation and employing the site and the pair approximations. We found active and nonactive steady states, and studied their stabilities. In one dimension, we found a reactive state only for a single value of the deposition rate ( $y_A = 1/2$ ) of the  $A$  monomer. The results we found, within the site and pair approximations, do not differ so much in the linear chain model. This one-dimensional model cannot be thought as being a particular case of the Bassler and Browne model [4], because here we prohibited the reaction between the monomers  $B$  and  $C$ . While the model of Bassler and Browne is symmetrical relative to all the possible reactions, our model is an asymmetrical one. Due to this fact, they found a clear reactive window in their phase diagram, while our model exhibits a reactive steady state only at the single point  $y_A = 1/2$ .

On the other hand, in two dimensions and in the pair approximation, we found a phase diagram, at the special value  $y_A = 1/2$ , showing active and nonactive states. The phase diagram, in the plane relative adsorption rate of the  $B$  and  $C$  monomers,  $r$ , versus probability of reaction of these two monomers with the  $A$  monomer,  $\beta$ , exhibits a first-order transition line. This phase transition does not appear in the site approximation, where, at  $y_A = 1/2$ , only an active state is present.

#### ACKNOWLEDGMENTS

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