Competing reaction model with many absorbing configurations

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We study a competitive reaction model between two monomers A and B on a linear lattice. We assume that monomer A can react with a nearest-neighbor monomer A or B, but reactions between monomers of type B are prohibited. We include in our model lateral interactions between monomers as well as the effects of temperature of the catalyst. The model is considered in the adsorption controlled limit, where the reaction rate is infinitely larger than the adsorption rate of the monomers. We employ site and pair mean-field approximations as well as static Monte Carlo simulations. We determine the phase diagram of the model in the plane y_A versus temperature, where y_A is the probability that a monomer of the type A arrives at the surface. This phase diagram shows regions of active and absorbing states separated by a line of continuous phase transitions. Despite the absorbing state of the model to be strongly dependent on temperature, we show that the static critical exponents of the model belong to the same universality class of the directed percolation.

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I. INTRODUCTION

Phase transitions in systems out of the thermodynamic equilibrium are very common in many branches of science. These transitions are obtained when some external parameters change, such as temperature, pressure, electric, or magnetic fields. Unfortunately, we do not have until now a successful theory to account for these problems as in the equilibrium statistical physics. The detailed balance condition fails in treating nonequilibrium states and the distribution of states according to the Boltzmann weight factor cannot be employed. Despite this fundamental difficult, the methods developed in the study of phase transitions in equilibrium statistical physics are usually extended to throw some light in the behavior of the nonequilibrium problems.

A particularly interesting class of problems that have been studied exhibits phase transitions from active states, where large scale spatial and temporal fluctuations are present, to inactive states, where fluctuations are completely absent. The simplest example of this type of phase transition is the contact process that models the spreading of a disease [1,2], where the external parameter that drives the system to an inactive (absorbing) state is the rate of change of healthy individuals into unhealthy ones. There are other examples of transitions to an absorbing state: surface catalytic reaction models [3], transition to turbulence [4], traffic flow [5], etc.

The relevant universality class associated to these transitions is the directed percolation (DP) [6,7]. The experimental determination of the critical exponents in the phase transition to an absorbing state is a difficult task, because in real systems there are always small fluctuations due to the presence of impurities and other inhomogeneities [8]. Recently, Kazumasa *et al.* [9] observed a phase transition between different turbulent states in nematic liquid crystals and showed that the set of static critical exponents is in full agreement with those defining the DP in 2+1 dimensions.

Catalytic reaction models are a class of nonequilibrium systems that exhibit a phase transition into absorbing states. Besides the mentioned Ziff, Gulari, and Barshad model [3], there are in literature many interesting examples of the catalytic reaction models [10–13]. In this work we focus our attention on a particular catalytic reaction model with competitive reactions between monomers of types *A* and *B*: the autocatalytic reaction $A+A \rightarrow A_2$ [14], and the simple monomer-monomer reaction $A+B \rightarrow AB$ [15]. This competitive reaction model was studied through dynamical mean-field approximation and Monte Carlo simulation [16,17], and the critical exponents of the model are in the same universality class of the directed percolation. The absorbing state is unique with the lattice completely poisoned by monomers of type *B*.

In order to allow the presence of phase transitions to multiple absorbing configurations we include in our model lateral interactions between nearest-neighbor adsorbed species [18-22] as well as thermal effects [23]. We determine the phase diagram of the model in the plane temperature versus partial pressure of monomers A in the gas phase. Depending on the temperature and partial pressure of the monomers of type A, the resulting absorbing state is a poisoned state with different concentrations of the A and B species. Despite this fact, the static critical exponents of the model are in the same universality class of the DP. This fact is interesting, because one of the premises of the DP assumption [6,7] is the uniqueness of the absorbing state. This paper is organized as follows: in the next section we describe the model and the mean-field calculations in the site and pair approximations. In Sec. III we present our Monte Carlo simulations along with finite-size scaling arguments to determine the critical behavior of the model. Finally, in the last section we present our conclusions.

II. INTERACTING REACTION MODEL

The system we are considering consists of two reacting models, the autocatalytic reaction $A+A \rightarrow A_2$ [14], and the simple monomer-monomer reaction $A+B \rightarrow AB$ [15] on a one-dimensional substrate. Monomers *A* and *B* arrive at the substrate, which is in contact with an infinite reservoir of monomers, with probabilities y_A and y_B , respectively, where $y_A+y_B=1$. These probabilities are related to the partial pres-

sure of the gases inside the reservoir. Each monomer can occupy only a single vacant site in the lattice and reactions can occur only if monomers are nearest neighbors in the lattice. The basic steps to describe the reactions are the following:

(i) $A_{(g)} + v \rightarrow A_{(a)}$, (ii) $B_{(g)} + v \rightarrow B_{(a)}$, (iii) $A_{(a)} + A_{(a)} \rightarrow A_{2(g)}$, and (iv) $A_{(a)} + B_{(a)} \rightarrow AB_{(g)}$.

where labels (g) and (a) denote a monomer in the gaseous and in the adsorbed phases, respectively. The first two steps describe the adsorption of the species on the substrate and the symbol v represents a vacant site. Steps (iii) and (iv) describe the possible reactions between adsorbed monomers occupying nearest-neighbor sites. As we will define next, the probability of a reaction between monomers depend on temperature and magnitude of the lateral interactions. Immediately after a reaction, A_2 or AB molecules leave the catalyst and a pair of nearest-neighbor vacant sites is generated on the substrate. We disregard in this model any type of diffusion of the species over the lattice.

This model was previously studied by Costa and Figueiredo [16], in the adsorption controlled limit, where the rate for the reactions is much larger than the rate for adsorption. There, always monomer A sees a nearest neighbor the reaction occurs without delay and the probability of A to react with another monomer A is the same as to react with monomer B. This model exhibits a unique absorbing state, which is characterized by a lattice completely poisoned by monomers of type B.

Besides the temperature of the catalyst, we also consider the lateral interactions between the adsorbed species. In the first place, for the adsorption process to occur, the existence of vacant sites on the lattice is not sufficient. Every time we try to deposit a new monomer on a vacant site of the lattice, we calculate the change in energy ΔE that this event would cause in the whole system. We assume that there is a repulsive energy $\epsilon(\epsilon > 0)$ between nearest-neighbor pairs of A monomers or A and B monomers, as well as, between monomers and catalyst. The interaction energy between two nearest-neighbor adsorbed monomers of type B is taken equal to zero. For any temperature, the probability that monomer B is adsorbed is one, provided that its nearest neighbors are vacant sites or occupied by monomers B. On the other hand, monomer A is always adsorbed if its nearestneighbor sites are both empty. Finally, thermal effects are taken into account only when we try to deposit a given monomer that can react with one of its nearest neighbors. The probability that the monomer is adsorbed on the substrate is chosen to be given by the Boltzmann-like weight factor

$$\alpha = \frac{2}{1 + e^{+\Delta E/k_B T}},\tag{1}$$

where the temperature *T* of the substrate is measured in units of ε/k_B , and k_B is the Boltzmann constant. The possible energy changes due to the deposition of a monomer that can react are $Avv \rightarrow AAv$, $Avv \rightarrow ABv$ and $AvB \rightarrow ABB$, for which $\Delta E = 2\varepsilon$, where v represents an empty site. For the transitions $AvA \rightarrow AAA$, $AvB \rightarrow AAB$, $AvA \rightarrow ABA$ and BvB $\rightarrow BAB$, we have $\Delta E=3\varepsilon$. Then, due to the interaction energy parameters of our model, we always have $\Delta E > 0$, which gives $\alpha \rightarrow 0$ when $T \rightarrow 0$ and $\alpha \rightarrow 1$ at high temperatures. Factor 2 in the last equation is to assure that probabilities are in the range from 0 to 1. If the monomer is not adsorbed, it reacts with probability $(1-\alpha)$. This choice recovers, in the limit $T \rightarrow 0$, the model considered by Costa and Figueiredo [16] where the absorbing state is unique, corresponding to a lattice completely filled with monomers B. In that model, it is not possible for a monomer of type A to stay adsorbed in the presence of another A or B monomers, and Eq. (1) for $T \rightarrow 0$ is consistent with an absorbing state with only B monomers in the lattice. As will see next, at a finite temperature, it is possible to find absorbing states with different concentrations of monomers A and B. Also, in the present model, the order parameter is the fraction of vacant sites in the lattice.

This study is not meant to describe realistic catalytic models. Our main purpose is to show that systems with a multitude of absorbing configurations can belong to DP universality class. This is not a trivial result because due to the thermal effects, when temperature is changed from very low to very high values, we found a continuous set of absorbing states. Concerning the values of the interaction energy between pairs of particles, and particles and catalyst, we would like to say that by using different figures for these energies, we only change the location of the line of critical points; however, the critical exponents, as is well known from the renormalization-group theory, are not affected by the magnitude of the interaction parameters.

A. Mean-field site approximation

In this approximation we disregard the correlations between nearest-neighbor sites. We define the densities p_i for each one of the species, that is, *i* can represent monomers *A* and *B* or the vacant sites *v*. The system is considered translationally invariant and the densities are normalized, $p_v + p_B$ $+p_A=1$. In order to write the master equation for the time evolution of these densities, we need to determine the transition probabilities corresponding to all possible processes concerning this competitive reaction model. We summarize them by the following five steps:

(1)
$$T_1:A+v \to A_{(a)}$$
,
(2) $T_2:A+v \to A_2\uparrow +2v$,
(3) $T_3:A+v \to AB\uparrow +2v$,
(4) $T_4:B+v \to B_{(a)}$, and
(5) $T_5:B+v \to AB\uparrow +2v$.

For instance, the rate corresponding to the second step is given by

$$T_2 = y_a p_v [2p_v p_A (1 - \alpha_1) + (p_A^2 + p_A p_B)(1 - \alpha_2)]$$

where $\alpha_{1,2} = 2[1 + e^{+\Delta E_{1,2}/k_B T}]^{-1}$, and $\Delta E_{1,2} = 2\varepsilon$, 3ε

The gain and loss equations for the densities of monomers A and B can be written in the form

$\frac{From}{To\downarrow}$	<i>v</i> – <i>v</i>	v-A	v-B	B-B	B-A	A - A
<i>v</i> - <i>v</i>	×	T_3	T_4	×	×	×
v - A	T_1	×	×	×	T_9	T_{10}
B-v	T_2	×	×	T_6	T_{11}	×
B-B	×	×	T_5	×	×	×
B-A	×	T_7	T_{12}	×	×	×
A - A	×	T_8	×	×	×	×

TABLE I. Possible transitions among different configurations of pairs of nearest neighbors in the lattice.

$$\frac{d}{dt}p_A = T_1 - T_2 - T_5$$
 (2)

and

$$\frac{d}{dt}p_B = T_4 - T_3. \tag{3}$$

The equation for the order parameter of the system p_v comes from the constraint $p_v+p_A+p_B=1$. We solve this system of equations by the fourth-order Runge-Kutta method with the initial condition $p_v=1$, i.e., empty lattice. We look for the stationary states of the model as a function of temperature and partial pressure of monomers *A* in the gaseous phase. In Sec. III we present the phase diagram for this model in this approximation along with the results from pair approximation and Monte Carlo simulations.

B. Mean-field pair approximation

This is the simplest approximation beyond the one-site approximation. Here, we introduce the correlation between two nearest-neighbor sites of the lattice. We introduce conditional probability P(i|j), which is the probability that a given site to be of type *i*, given that one of its nearest neighbors is of type *j*. Therefore, we can write for the probability of a nearest-neighbor pair (ij) in the lattice

$$p_{ii} = p_i P(i|j). \tag{4}$$

The pair probabilities are related to the densities of monomers A and B and to the fraction of vacant sites by the relation

$$p_j = \sum_i p_{ij}.$$
 (5)

Pairs (ij) and (ji) occur with the same probability in the lattice and this model allows the presence of all pairs of nearest-neighbor sites in the lattice. Therefore, we can write for the densities

$$p_A = p_{vA} + p_{BA} + p_{AA},$$

$$p_B = p_{vB} + p_{BB} + p_{BA},$$

$$p_v = p_{vv} + p_{vA} + p_{vB}.$$
(6)

In this approximation [24] we assume that the probability for a larger cluster is written as a product of nearest-neighbor pair probabilities, and that the system is translationally invariant. As we have made in the site approximation, we need to consider the change in energy after we try to deposit a given monomer on the substrate. All the possible transitions between pairs of nearest neighbors in the lattice can be seen in the Table I. In this table the \times means that the transition is not possible. For instance, the transition from pair *BA* to *Bv* occurs with a rate transition T_{11} every time monomer *A* or *B* arrives at a empty site in the vicinity of monomer *A* of this central pair. Then, we can write the following expression for the transition rate T_{11} :

$$T_{11} = y_a p_{BA} \left(\frac{p_{vA}}{p_A} \right) \left[\left(\frac{p_{vv}}{p_v} \right) (1 - \alpha_1) + \frac{1}{2} \left(\frac{p_{vA} + p_{vB}}{p_v} \right) (1 - \alpha_2) \right] + y_b p_{BA} \left(\frac{p_{vA}}{p_A} \right) \left[\left(\frac{p_{vB} + p_{vv}}{p_v} \right) (1 - \alpha_1) + \frac{1}{2} \left(\frac{p_{vA}}{p_v} \right) (1 - \alpha_2) \right],$$
(7)

where the probabilities α_1 and α_2 are the same used in the site approximation. After we calculate the other transition rates of Table I, we can establish the set of gain-loss equations for the pair densities

$$\frac{d}{dt}p_{vB} = T_2 + T_6 + T_{11} - T_4 - T_5 - T_{12},$$
(8)

$$\frac{d}{dt}p_{\nu A} = T_1 + T_9 + T_{10} - T_3 - T_7 - T_8,$$
(9)

$$\frac{d}{dt}p_{vv} = 2(T_3 + T_4 - T_1 - T_2), \tag{10}$$

$$\frac{d}{dt}p_{BA} = T_7 + T_{12} - T_9 - T_{11}, \tag{11}$$

$$\frac{d}{dt}p_{AA} = 2(T_8 - T_{10}), \tag{12}$$

$$\frac{d}{dt}p_{BB} = 2(T_5 - T_6).$$
(13)

This last equation can also be obtained from the constraint



FIG. 1. Order parameter p_v as a function of y_A in the site and pair mean-field approximations. Monte Carlo simulations are also shown for four lattice sizes *L*. The temperature is T=0.5, and it is measured in units of ϵ/k_B .

$$p_{BB} = 1 - p_{AA} - p_{vv} - 2(p_{vA} + p_{BA} + p_{vB}), \qquad (14)$$

which comes from Eq. (6) and from the normalization for the densities $p_A + p_B + p_v = 1$. Again, we solve this set of simultaneous equations by the fourth-order Runge-Kutta method starting with the initial condition $p_v = 1$. The stationary states of the model are determined as a function of temperature and partial pressure of monomers *A* in their gaseous phase y_A and the results will be present in the next section.

C. Monte Carlo simulations

We have determined the phase diagram and the critical behavior of the model through Monte Carlo simulations. We consider a linear lattice of size L, with L ranging from 256 to 16 384. We employ periodic boundary conditions and we start our simulations with an empty lattice. We applied a continuous time algorithm in order to save time. In this way, instead of looking for an empty site in the whole lattice, we restrict our search to the sites of a list of empty sites at each instant of time. This is very efficient because close to the absorbing states the number of empty sites is very small.

For each value of temperature T and partial pressure y_A of the monomers of type A in the gaseous phase the algorithm reads: (i) a site is randomly chosen from the list of empty sites in the lattice, and we count the number of monomers A



FIG. 2. The same legend as in Fig. 1 but for T=1.3.



FIG. 3. Phase diagram of the model in the plane y_A versus *T*. Monte Carlo simulations (squares), pair mean-field approximation (inverted triangles) and site mean-field approximation (circles). Each critical line separates the active phase (top) from the absorbing phase (down).

and B that are its nearest neighbors; (ii) with probability y_A , monomer A will be adsorbed in the selected site if there is no A nor B monomer in its neighborhood, and in this case the list of empty sites decreases by one. If there is at least one monomer A or B in its neighborhood, then we take the following action: we calculate the probability α , Eq. (1), and select a random number between 0 and 1; if α is larger than the random number then monomer A is adsorbed; and if not, it reacts and leaves the lattice, increasing the number of empty sites of the list by one unit. Similarly, with probability $y_B = 1 - y_A$, monomer B will be adsorbed in the lattice unless there is at least one monomer of type A in its neighborhood. In this case we also need to determine the probability α in order to know if monomer B will be adsorbed in the selected site. We consider equal reactivities for both species, that is, if we have as nearest neighbors of monomer A, monomer B, and another monomer A, the reaction, if it occurs, is chosen randomly between the two possible paths.

After the system reaches the stationary state, we start measuring the densities of both species, p_A and p_B , as well as the density of vacant sites in the lattice, p_v . We would like to mention that only the surviving configurations, which did not



FIG. 4. Coverages of species A and B, in the Monte Carlo simulations, as a function of temperature for which we have the phase transition from the active to the absorbing state. The concentration of empty sites is zero.



FIG. 5. Coverages of species A and B and of the empty sites, in the Monte Carlo simulations, as a function of y_A at T=1.35.

enter into the absorbing state, are used to calculate the average values of the densities. We show in Figs. 1 and 2 the mean-field results along with the Monte Carlo simulations, for the order parameter p_v as a function of y_A for two selected values of temperature. The transition from the active to the absorbing state is always continuous for any value of temperature. At low temperatures, as in Fig. 1, the agreement between mean-field results and Monte Carlo simulations is good only far away of the critical point. As to be expected, at the critical point, pair and site approximations underestimate the critical value of the partial pressure. However, when we increase the temperature. Monte Carlo results are completely different from the mean-field calculations as we can see in Fig. 2 for T=1.3 for all values of the parameter y_A . At high temperatures, the probability of monomer A to become adsorbed is very large, and the partial pressure of A monomers needs to increase in order to have an active state in the lattice.

Figure 3 displays the phase diagram in the plane y_A versus T in the site and pair mean-field calculations as well as in the Monte Carlo simulations. For each value of temperature, there is a critical value of the partial pressure y_A , below which the lattice is completely poisoned by monomers A and B. However, there is a maximum value of temperature to have an active state in the lattice. For instance, in the site



FIG. 7. The same legend as in Fig. 6 but for T=1.3 and straight line (stars).

approximation this temperature is T=2.70, in the pair approximation T=1.83, and in the simulations we have T=1.347. We show in Fig. 4 the concentration of species A and B as a function of temperature at the corresponding critical point.

While at T=0 the critical point is characterized by a lattice completely filled with *B* monomers, at temperatures larger than T=1.35 the absorbing state is formed only by monomers of type *A*. However, at intermediate temperatures, the absorbing state is a mixture of the species. In fact, we observe a continuum of absorbing states from T=0 to T=1.35. Figure 5 shows the coverages of species as a function of the critical partial pressure for T=1.35, which is a little above the maximum temperature observed in the Monte Carlo simulations. Again, we see that the absorbing state is a continuum of configurations ranging from a pure *B* state at $y_A \approx 0$ to a pure *A* state at $y_A \approx 1$.

We have shown in Fig. 3 the phase diagram of the model, where continuous phase transitions occur from the active to absorbing states. To find the critical exponents of the model, we choose some points of the phase diagram, corresponding to different temperatures, and we consider the behavior of the order parameter, which in our case is the fraction of empty sites, p_v , in the neighborhood of these points. We performed a finite-size scaling analysis for the order parameter [25]. We assume that it is a generalized homogeneous



FIG. 6. Log-log plot of the order parameter p_v versus the system size *L* for some selected values of y_A as indicated in the figure. The temperature is T=0.5 and the straight line (triangles) gives the critical value of the parameter y_A .



FIG. 8. Collapse of the data points of Fig. 6. The figure is a log-log plot of $p_v L^{\beta/\nu_{\perp}}$ versus $(y_A - y_{A_c}) L^{1/\nu_{\perp}}$. The best collapse was found with the values $\beta = 0.272(1)$ and $\nu_{\perp} = 1.083(8)$.



FIG. 9. Collapse of the data points of Fig. 7. The figure is a log-log plot of $p_v L^{\beta/\nu_\perp}$ versus $(y_A - y_{A_c}) L^{1/\nu_\perp}$. The best collapse was found with the values $\beta = 0.269(3)$ and $\nu_\perp = 1.087(4)$.

function of variables *L*, the linear size of the system, and $\Delta = y_A - y_{A_c}$, which measures the distance from the critical point y_{A_c} ,

$$p_{\nu}(\Delta, L) \propto L^{-\beta/\nu_{\perp}} f(\Delta L^{1/\nu_{\perp}}), \qquad (15)$$

where β and ν_{\perp} are the order-parameter critical exponent and the critical exponent associated with the spatial correlation length, respectively. The scaling function $f(x) \propto x^{\beta}$ holds for large values of x. At the critical point where $\Delta = 0$, we can write that

$$p_v \propto L^{-\beta/\nu_\perp},\tag{16}$$

and we can find the ratio β/ν_{\perp} from the straight line behavior of the log-log plots of p_v versus *L*. We show in Figs. 6 and 7 the behavior of p_v versus *L* for the temperatures *T* =0.5 and *T*=1.3, respectively. The central curve in Fig. 6 gives the critical value y_{A_c} =0.6432(1) and for the ratio β/ν_{\perp} =0.256 89(6). Analogously, from Fig. 7, we found the critical value y_{A_c} =0.868 55(10) and the ratio β/ν_{\perp} =0.248 86(6). In Figs. 6 and 7 as well as in Figs. 8 and 9, we considered the following values of *L*: 256, 512, 1024, 2048, 4096, 8192, and 16 384, although we did not include all the points in these plots in order to leave these figures more clear. However, all seven lattices were used in the calculation of the critical point and the related critical exponents.

We can also estimate the critical exponents by collapsing the data points of Figs. 6 and 7 obtained for four different lattice sizes. From the log-log plot of $p_n L^{\beta/\nu_\perp}$ versus $(y_A - y_{A_c})L^{1/\nu_{\perp}}$ we can tune the values of β and ν_{\perp} in order to have the data points in the same curve. For large values of the argument of the function f in Eq. (15), the data should fall on a straight line with slope β . Figure 8 displays the collapse of the data for T=0.5, while Fig. 9 shows the collapse for T=1.3. For T=0.5, the best results we find are β =0.272(1) and $\nu_{\perp}=1.083(8)$, and for T=1.3 we find β =0.269(3) and $\nu_{\perp}=1.087(4)$. In Table II, we summarize the results for some selected temperatures as well as the results found earlier at T=0 [16], and the best values for the DP universality class [26]. Despite the system to exhibit many different absorbing states, which depend on temperature, our exponents are in close agreement with those of the Directed Percolation.

III. CONCLUSIONS

We have studied in this work a one-dimensional model of competitive reactions where we considered thermal effects and lateral interactions between nearest-neighbor monomers. In our analysis we have employed mean-field calculations at the site and pair approximations as well as Monte Carlo simulations. The model exhibits continuous phase transitions between active and absorbing states. We determined the phase diagram of the model in the plane temperature versus partial pressure of one of the reactants. We have found a continuum of absorbing states when we change the temperature from low to high values. Although we have determined a continuum of absorbing states as a function of temperature, the critical exponents β and ν associated to the transition from the active to absorbing states do not depend on temperature. The values we have found for these critical exponents are in the DP universality class. This fact does no contradict the DP conjecture advanced by Grassberger [7], where continuum phase transitions to an absorbing state, with local evolution rules and not conserved order parameter, belong to the directed percolation universality class. There are other examples in the literature of multiple absorbing configurations but with a unique absorbing state for which the order parameter is zero. This is the case observed in the pair contact process [27] and in some models in catalysis [28-30]. A word of caution must be said about the static and dynamical critical properties of the models with multiple absorbing states. For instance, the one-dimensional pair contact process and the dimer reaction models belong to the DP

TABLE II. Critical exponents of the order parameter (β) and of the correlation length (ν_{\perp}) for some selected temperatures.

Exponent	$T=0^{a}$	<i>T</i> =0.5	<i>T</i> =0.9	T=1.3	<i>T</i> =1.33	DP ^b
β	0.27(2)	0.272(8)	0.276(8)	0.269(7)	0.271(8)	0.276486(8)
$ u_{\perp}$	1.07(3)	1.083(8)	1.088(3)	1.087(4)	1.088(7)	1.096854(4)
^a Reference [16].					

^bReference [26].

universality class regarding their static critical behavior [27,31]. However, the dynamical critical properties of both models are nonuniversal, the exponents depending upon the nature of the initial conditions [31]. In order to describe this complex behavior, a generalized scaling theory was devised where the dynamic critical exponents can change according to the initial configurations [32].

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