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Study of the one-dimensional off-lattice hot monomer reaction model

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Abstract. Hot monomers are particles having a transient mobility (a ballistic flight) prior to being definitely absorbed on a surface. After arriving at a surface, the excess energy coming from the kinetic energy in the gas phase is dissipated through degrees of freedom parallel to the surface plane. In this paper we study the hot monomer–monomer adsorption–reaction process on a continuum (off-lattice) one-dimensional space by means of Monte Carlo simulation. The system exhibits a second-order irreversible phase transition between reactive and saturated (absorbing) phases which belong to the directed percolation (DP) universality class. This result is interpreted by means of a coarse-grained Langevin description which allows us to extend the DP conjecture to transitions occurring in continuous media.

1. Introduction

Interacting particle systems are relevant to wide-ranging phenomena in physics, chemistry, biophysics, ecology, etc. The concept of ‘particles’ is used in a broad sense, that is ‘particles’ can be atoms, molecules, spins, individuals, etc, and whilst attention is drawn to the interactions among particles no attempt is made to achieve a detailed description (e.g. quantum mechanical) of the particle itself. Therefore, due to interactions, the occurrence of complex behaviour, such as phase transitions, self-organization, chaos, bistability, etc, may be observed [1].

Within this context, an active field of research is the study of far-from-equilibrium reaction systems [2, 3]. Irreversible phase transitions (IPTs) between active regimes (where reactions are sustained) and absorbing states (where reactions are no longer possible) have been reported in a great variety of models such as the Ziff, Gulari and Barshad (ZGB) model for the catalytic oxidation of CO [2], the dimer–dimer model [4], the contact process [5, 6], forest-fire models [7], etc (for a recent review see e.g. [3]). According to the Janssen–Grassberger conjecture [8, 9], irreversible reaction systems that exhibit a phase transition to a single absorbing state characterized by a scalar order parameter belong to the directed percolation (DP) universality class. This conjecture, stated a long time ago for unique absorbing states, has been further generalized for the cases where such states are non-unique [6, 10]. A special case corresponds to non-equilibrium systems where, provided an IPT exists, there is in addition a local or global conservation of particles of modulo two, such as the branching and annihilating random walks with an even number of offsprings [11, 12]. In these cases a new universality class emerges, commonly called the parity conserving (PC) class, which is due to the existence of two statistically equivalent absorbing states at the critical point [13]. However, global

conservation of particles of modulo two may also lead to exponents in the PC class only when local spontaneous annihilation ($1X \rightarrow 0$) is highly inhibited. Then, at a coarse-grained level, the relevant surviving processes are those conserving parity. In other words, parity conservation can be restored at a coarse-grained level [14, 15]. A nice example where global parity conservation still leads to DP exponents is given by Inui *et al* [16]. It is clear in this case that spontaneous annihilation must be taken into account.

IPTs are studied largely by means of Monte Carlo simulations and mean field approaches. Recent developments of field-theoretic renormalization group techniques have provided a new theoretical framework where non-equilibrium phase transitions can be studied [17]. These techniques are able to identify the relevant processes in a given universality class although the quantitative predictions are still poor.

So far, most of the simulations have been performed using *discrete* lattices where each particle fills a single site on the lattice and neighbouring particles may react with a certain probability. In contrast, our knowledge of the behaviour of irreversible reaction systems in continuous media is rather poor. In order to stress some dramatic differences that may arise for a reaction system when it is simulated off-lattice, let us consider the simplest case of the $B + B \rightarrow 0$ irreversible reaction which proceeds according to the Langmuir–Hinshelwood mechanism:



where g and a refer to the gas and adsorbed phases, respectively, while S represents a site on the surface. At first, we assume that B species adsorbed on nearest-neighbour sites react with unitary probability ($P_r = 1$). If we used a discrete lattice, reactions would be sustained indefinitely, i.e. the system could not irreversibly evolve into an absorbing state. However, considering a continuous media, the random adsorption of B particles of finite size σ causes the formation of several interparticle gaps of size smaller than σ . So, in the infinite time limit ($t \rightarrow \infty$) the sample becomes imperfectly covered by B species separated by small interparticle gaps. Reaction is no longer possible and the system becomes irreversibly trapped into an absorbing state (infinitely degenerated). The maximum jamming coverage attained in one dimension is $\Theta_j \approx 0.74759$ ($P_r = 0$), which corresponds to the so-called car parking problem [18, 19].

In this paper we show that by introducing the adsorption of species with transient mobility in a continuous one-dimensional (1D) medium, it is possible to open a window where reactions are sustained. However, by tuning the external parameter which controls the transient mobility of particles it is possible to irreversibly drive the system into an absorbing state.

It should be mentioned that the study of reactions of atoms in the gas phase possessing thermal energy with adsorbed atomic CO species on metal and semiconductor surfaces is a topic of current interest. In contrast to thermally activated reactions among adsorbed species, i.e., the so-called Langmuir–Hinshelwood mechanism, these kind of reactions take place under far-from-equilibrium conditions. Consequently, the determination of the underlying mechanism as well as the understanding of the dynamic behaviour is challenging work. Within this context, very recently Kim *et al* [20] have reported experimental studies of the reaction of hot H atoms with adsorbed D atoms (for further experimental works see the references in [20]).

It should be noted that from the theoretical point of view a number of related models for random sequential adsorption with diffusion [21, 22] and desorption [23] have also been proposed and studied (for a review see also [18]). However, interest in such studies is addressed to the asymptotic approach to the jammed state. In contrast, in this paper our interest is focused

on the irreversible critical behaviour of a reactive system.

So this paper is devoted to the characterization of such IPT in continuous media and it is organized as follows: section 2 gives the description of the model and the simulation technique. In section 3 we discuss the results while conclusions and remarks are presented in section 4.

2. The model and the Monte Carlo simulation method

In this paper, we study a 1D off-lattice adsorption–reaction model in which particles of size σ undergo a ballistic flight just after deposition on the substrate. The system evolves in time under the following local rules. (i) A position x is randomly selected on the substrate. If the interval $[x - \sigma/2, x + \sigma/2]$ is empty, then the adsorption trial is successful, otherwise it is rejected. So, it is clear that double occupancy of positions is forbidden. (ii) Right after a successful adsorption trial, a random direction is selected (left or right). Then, the particle undergoes a ballistic flight in the previously selected direction up to a distance R from the adsorption position x , provided that no other already deposited particle is found along the way. (iii) If during the flight one particle hits another previously adsorbed particle which is already at rest on the substrate, the following alternatives can occur: (1) the annihilation ($B + B \rightarrow 0$) occurs with probability P_r . Then, both particles react and leave the system. (2) Particles do not react (with probability $(1 - P_r)$), and the flying particle is frozen at the collision point.

The ballistic flight mimics ‘hot monomer’ adsorption, allowing the incoming particle to transform its energy into degrees of freedom parallel to the substratum. The length of the flight R is finite in order to account for frictional dissipation. The model has two externally tunable parameters, namely R and P_r . For $P_r = 0$ one recovers the ‘hot monomer’ random sequential adsorption model [24] while for $R = 0$ and $P_r = 0$ one has the 1D car parking problem [19].

In order to simulate a continuous medium on a digital computer, one actually considers a discrete lattice. However, each site of size σ is subdivided into 2^{64} different adsorption positions. This high degree of discretization has provided excellent results when compared with the exact analytic solution of a related problem [24].

Preliminary results show that the system can undergo continuous IPTs between a stationary reactive state and an absorbing state without reactions when varying the parameters. This can easily be tested by considering the case $P_r = 1$ and $R = 0$ ($R > 1$) which gives an absorbing (reactive) state, respectively. It should be pointed out that continuous IPTs are dominated by fluctuations. Consequently, in a finite system and close to the critical point, the stationary state of the reactive phase can irreversibly evolve into the saturated state (absorbing state). Due to this circumstance, the precise determination of both critical points and critical exponents is rather difficult. However, this shortcoming can be avoided by performing an epidemic analysis. For this purpose one starts, at $t = 0$, with a configuration close to one of the absorbing states. It is clear that different absorbing states will normally differ in the density of monomers. It should be pointed out that the dynamical critical behaviour of systems with infinitely many absorbing configurations is expected to depend upon the initial density of inactive particles [6, 25]. However, static critical behaviour appears to be independent of it. From the set of possible initial densities, a value ρ_n is particularly important, namely the stationary density of inactive particles which results after the natural evolution of the system in the subcritical region has finished. The value ρ_n is relevant, since it is only for this value that the natural dynamical critical behaviour emerges. Preliminary simulation results show that ρ_n depends on the parameter P_r , but their values have not been included in this work for the sake of space. The dependence of the critical behaviour on the set of initial densities is the subject of an ongoing investigation.

Consequently, the initial state for the epidemic analysis is generated by the evolution of

the system very close to the critical point until poisoning is achieved. After generating this stationary poisoned state, we remove one or two particles from the middle of the system in order to create a small active area where adsorption is now possible. It should be noted that an empty area is considered to be active if it is longer than or equal to σ . Then, the time evolution of the system is analysed by measuring the following properties: (i) the average amount of active area at time t , $A(t)$; (ii) the survival probability of the active area at time t , $P_s(t)$; and (iii) the average distance over which the active area has spread at time t , $D(t)$. Finite-size effects are absent because the system is taken large enough to avoid the presence of active area at the boundaries. For this purpose a sample of $10^4\sigma$ is enough. Averages are taken over 10^5 to 10^6 different samples. Near the critical point, the amount of active area is often very small. Then, we improve the efficiency of the algorithm by keeping a list of the positions where there is active area. Time is incremented by $1/a(t)$, where $a(t)$ is the amount of active area at time t . The time evolution of the active area is monitored up to $t = 10^5$. At criticality, the following scaling behaviour holds:

$$A(t) \propto t^\eta \quad (2.1)$$

$$P_s(t) \propto t^{-\delta} \quad (2.2)$$

and

$$D(t) \propto t^{z/2} \quad (2.3)$$

where δ , η and z are *dynamic* exponents.

3. Results and discussion

Preliminary simulations show that for $P_r = 1$ it is possible to achieve a stationary reactive state in the large- R limit (i.e. for $R \geq 2$) while in the opposite limit ($R \leq 1.5$) the system becomes irreversibly saturated by B species. In order to obtain a quantitative description of the IPT we have performed epidemic studies around the critical edge. Figures 1(a)–(c) show log–log plots of A , P_s and D versus the time t , obtained for different parameter values. The three plots exhibit a power law behaviour which is the signature of a critical state. Using smaller (greater) R values we observe slight upward (downward) deviations in the three plots which indicate supercritical (subcritical) behaviour (these results are not shown for the sake of clarity). Then, the critical exponents obtained by regressions are

$$\eta = 0.308 \pm 0.004 \quad \delta = 0.165 \pm 0.003 \quad z/2 = 0.625 \pm 0.002. \quad (3.1)$$

These values are in excellent agreement with the exponents corresponding to the DP universality class in $1 + 1$ dimensions [8, 9]. Recently, extended series expansion calculations [26] have provided very accurate values for the DP critical exponents, namely

$$\eta = 0.313\ 68(4) \quad \delta = 0.159\ 47(3) \quad z/2 = 0.632\ 61(2). \quad (3.2)$$

Therefore, we conclude that the studied adsorption–reaction model on a *continuous medium* belongs to the DP universality class like many other systems already studied on *discrete lattices*. It should be noticed that the present model has infinitely many absorbing states, so as in the case of the dimer–dimer model [4] the DP conjecture holds for non-unique absorbing states [8, 10] at least as long as the absorbing states can be solely characterized by the vanishing of a single scalar order parameter.

We have also studied the case of imperfect reaction, i.e. $P_r < 1$. Figure 2 shows a plot of the phase diagram. The phase boundary curve was determined by means of an epidemic analysis, as is shown in figure 1. The obtained critical exponents are

$$\eta = 0.312 \pm 0.004 \quad \delta = 0.157 \pm 0.003 \quad z/2 = 0.631 \pm 0.001. \quad (3.3)$$

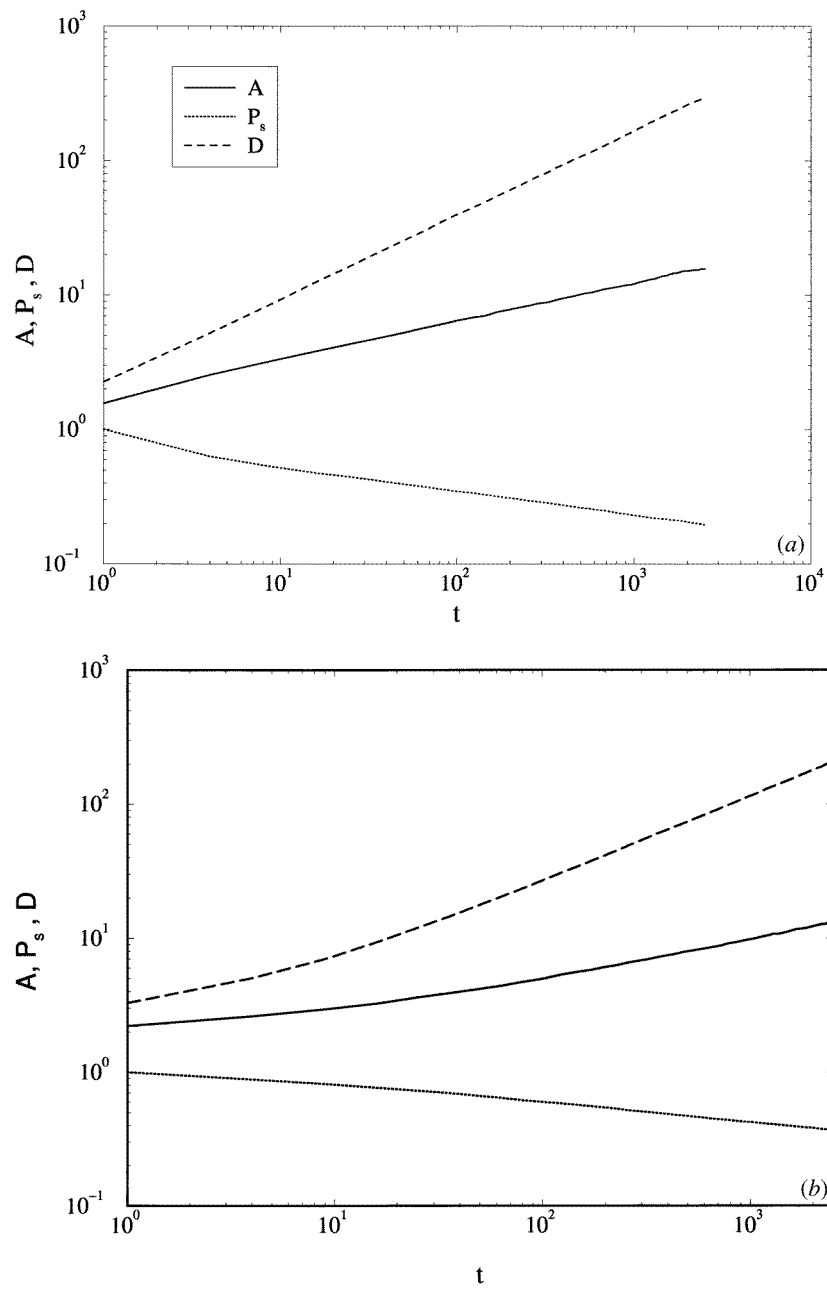


Figure 1. Log-log plots of the active area $A(t)$, the survival probability $P_s(t)$ and the epidemic diameter $D(t)$ versus t . Results are obtained for: (a) $P_r = 0.60$ and $R = 6.40$, (b) $P_r = 0.80$ and $R = 1.685$ and (c) $P_r = 1$ and $R = 0.853$. Averages are taken over (10^5) runs with different poisoned initial states.

Once again, these exponents are in good agreement with those corresponding to DP. Scanning the whole critical curve we obtain second-order IPTs that belong to the DP universality class. However, the special case $R \rightarrow \infty$ merits further discussion. For $P_r = 1$ ($P_r = 0$), the system

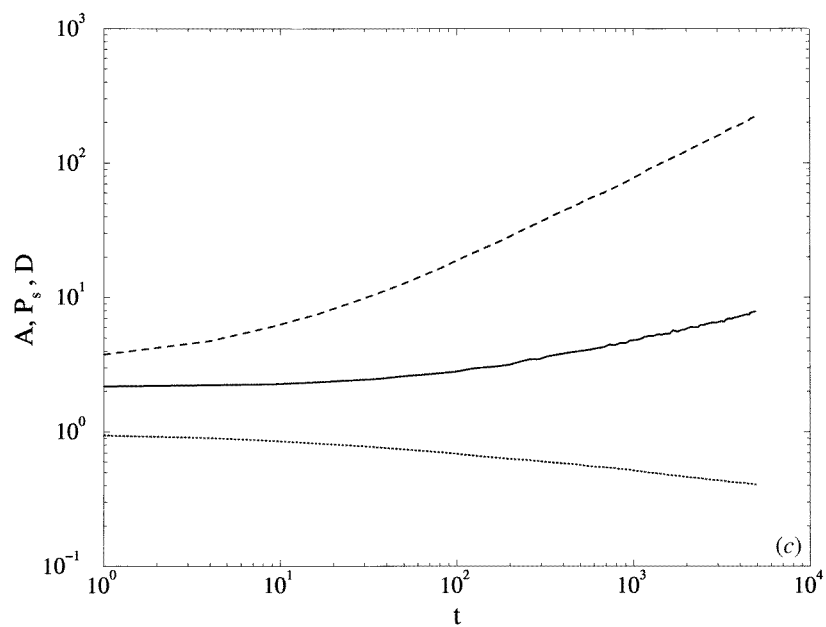


Figure 1. (Continued)

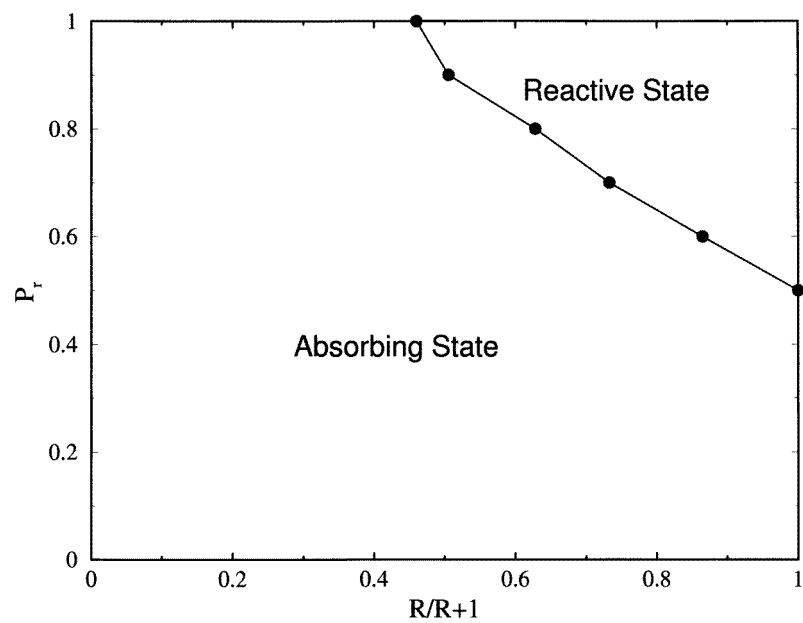


Figure 2. The phase diagram for the $B + B = 0$ reaction on the 1D continuum (off-lattice). The active and adsorbing states zones are shown on a P_r versus $R/(R + 1)$ plot. The extreme values of R in the diagram are 0.853 and ∞ for $P_r = 1$ and 0.5, respectively. According to figures 1 and 3 the point ($R = \infty$, $P_r = 0.5$) is the only one which does not correspond to the DP universality class.

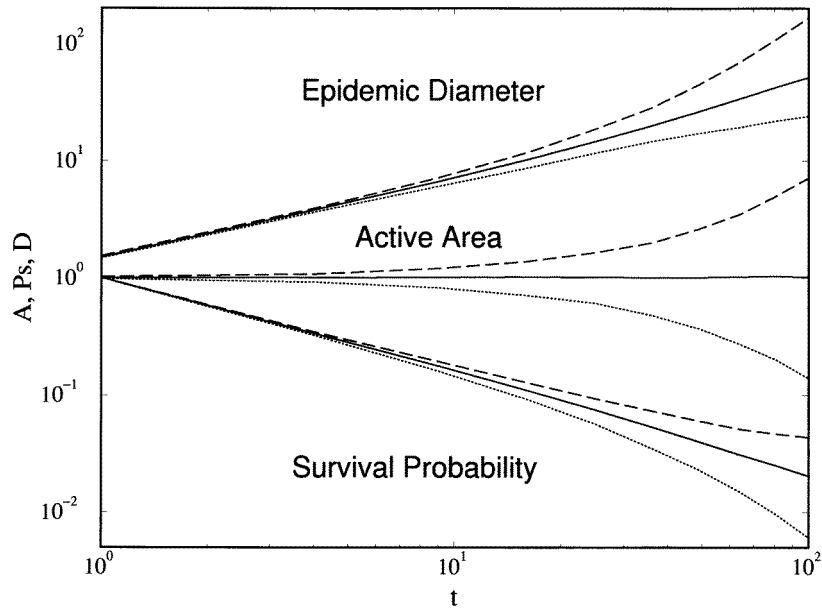


Figure 3. Log-log plot of the active area $A(t)$, the survival probability $P_s(t)$ and the epidemic diameter $D(t)$ versus t for the singular case with $R = \infty$. The solution of this case is exactly the one given by the mean field approximation. Dotted, full and dashed curves correspond to epidemic studies for $P_r < 0.5$, $P_r = 0.5$ and $P_r > 0.5$, respectively.

evolves towards a reactive (absorbing) state, respectively. Then, a transition is expected at some intermediate P_r value. In this case, the time evolution of the active area can be described by means of a mean field equation. In fact, the active area $A(t)$ will grow (decrease) proportionally to $A(t)P_r$ ($A(t)(1 - P_r)$), respectively; so

$$\frac{dA}{dt} = A(t)P_r - A(t)(1 - P_r) \tag{3.4}$$

which leads to

$$A(t) = A_0 e^{(2P_r - 1)t} \tag{3.5}$$

Therefore, $P_r = \frac{1}{2}$ is a critical value such that for $P_r > \frac{1}{2}$ ($P_r < \frac{1}{2}$) the active area will increase (decrease) exponentially in time, while just at criticality $A(t)$ will remain constant ($A(t) = A_0$), which is consistent with a mean field exponent $\eta_{MF} = 0$. The predicted behaviour is confirmed by means of simulations as is shown in figure 3. By means of linear regressions the following exponents are obtained for $P_r = \frac{1}{2}$:

$$\eta \approx 0.0 \quad \delta \approx 1.0 \quad z/2 \approx 1.0. \tag{3.6}$$

Then, our mean field estimate for η is in good agreement with the simulation results. Regrettably, we were unable to derive the mean field values for the remaining exponents.

We conclude that the particular point $P_r = \frac{1}{2}$, $R \rightarrow \infty$ is a first-order point (see figure 2) which is not in the DP class which characterizes the whole critical curve.

In the following, we give theoretical arguments by means of a coarse-grained Langevin description that supports the result concerning the universality class of the model. First, note that the normalized variables needed to characterize the configurations of the system are the amount of active area $a(x, t)$, the number of monomers in the system $n(x, t)$ and the amount

of inactive area $v(x, t)$. These variables are not independent since we have the constraint $a(x, t) + n(x, t) + v(x, t) = 1$. It is clear from the above discussion that the time evolution of the system ends when $a(x, t) = 0$. Since $a(x) \rightarrow 0$ at criticality, this quantity can be chosen as the order parameter of the system. Then, we will try to describe the time evolution of the system near the critical point by means of two coupled Langevin equations, for instance, one for $a(x, t)$ and the other for $n(x, t)$. Due to the nature of the absorbing configurations, each term of these equations must vanish when $a(x, t) \rightarrow 0$.

Let us consider the microscopic processes which are relevant to characterize the critical behaviour of the system. First of all, both diffusion of $a(x)$ and $n(x)$ can be interpreted as successive adsorption–reaction processes. Within a one-site description, both $n(x)$ and $a(x)$ will increase proportionally to $a(x)$. The reaction processes will contribute to the equations with a coupling term proportional to $a(x)n(x)$. It is also clear that monomer flights will introduce terms proportional to $a(x)^2$, $a(x)^3$, etc. Since only the lower-order terms are relevant for a renormalization group treatment [27], we just keep the term proportional to $a(x)^2$. Then, we can write down the following Langevin equations:

$$\partial n(x, t)/\partial t = k_1 \nabla^2 a(x, t) + k_2 a(x, t) - k_3 a(x, t)^2 - k_4 a(x, t)n(x, t) + \eta_1(x, t) \quad (3.7)$$

$$\partial a(x, t)/\partial t = u_1 \nabla^2 a(x, t) + u_2 a(x, t) - u_3 a(x, t)^2 - u_4 a(x, t)n(x, t) + \eta_2(x, t) \quad (3.8)$$

where $\eta_1(x, t)$ and $\eta_2(x, t)$ are uncorrelated noises proportional to $\sqrt{a(x, t)}$, k_i and u_i are coefficients. This system of coupled Langevin equations is similar to that obtained for the ‘pair contact process’ [5, 6] which is one of the prototype systems with multiple absorbing states. Muñoz *et al* [28] have shown that for large t the equation corresponding to the activity (equation (3.8) for the present model) reduces to the Langevin representation of DP. Then, our simulation results are consistent with the above-presented theoretical arguments. The same authors have also shown that systems with many available absorbing configurations display strong memory effects that may lead to anomalous scaling. In addition to this, Mendes *et al* [25] have proposed a generalized hyperscaling relation which has proved to be valid in systems with multiple absorbing configurations. Simulation results on several lattice models with infinitely many absorbing states support both theoretical arguments [6, 25, 29]. The role that initial states play in the temporal evolution of the present model is under investigation.

4. Conclusions and final remarks

A model for the irreversible adsorption–reaction of a single species on a continuous medium is studied by means of numerical simulations. We would like to stress and comment upon the following interesting features of the system: (i) in contrast to standard (reversible) transitions, non-equilibrium IPT can happen in one dimension. (ii) The studied adsorption–reaction model clearly shows interesting new effects that may arise when a process is modelled on a continuous medium. Since the system always reaches a stationary reactive state when simulated on a discrete lattice but a final poisoned state can be observed on a continuous one (e.g. for $P_r = 1$ and $R = 0$), one may expect a crossover behaviour when the ‘discretization degree’ of the surface is tuned from one extreme to the other. This can be achieved by considering the adsorption on *discrete lattices* of species of arbitrary length r , i.e. r -mers. We found that the reactive random sequential adsorption (RRSA) of dimers ($r = 2$) always leads to a reactive steady state, whose stationary coverage is close to $\Theta \approx 0.5$, and no poisoning is observed. However, the RRSA of trimers ($r = 3$) causes irreversible poisoning of the lattice with an average saturation coverage close to $\Theta_{ST} \approx 0.7$. In the case of dimers, two absorbing states

of the type

$$\begin{aligned} & \dots BBVBBVBBVBB \dots \\ & \dots VBBVBBVBBVBB \dots \end{aligned} \quad (4.1)$$

where V is an empty site, can be expected. So, during the RRSA the formation of several interfaces of the type $\dots BBVBBVBBVBB \dots$ takes place. Due to coarsening we expect that in the asymptotic limit ($t \rightarrow \infty$) both absorbing states will form two semi-infinite domains separated by an interface. The competition between these domains will keep the system in the reactive state for ever. Just by increasing $r = 2$ to 3, an infinite number of absorbing configurations appear in the system. So, the arguments developed above no longer hold and poisoning is observed. Consequently, an IPT is located between $r = 2$ and 3 and its precise location requires the study of the adsorption–reaction problem with particles of non-integer length. However, adsorption of r -mers of length $r = 2 + \epsilon$ would cause the argument of the two competing absorbing states to fail. Therefore, we expect that the critical size, i.e. ‘the discretization degree’ is 2. (iii) To our best knowledge, this is the first off-lattice model which exhibits a second-order IPT in the DP universality class. Consequently, this result once again supports the DP conjecture [8] which can now be generalized to off-lattice models with infinitely many absorbing states.

We expect that the interesting behaviour of the present simple reaction model will stimulate further work on irreversible transitions and critical phenomena on continuous media, a field that, to our best knowledge, remains almost unexplored.

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