Reactive dynamics on fractal sets: anomalous fluctuations and memory effects

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PACS. nn.mm.xx – 05.40.
PACS. nn.mm.xx – 05.45.Df.
PACS. nn.mm.xx – 82.20.

Abstract. – We study the effect of fractal initial conditions in closed reactive systems in the cases of both mobile and immobile reactants. For the reaction \( A + A \rightarrow A \), in the absence of diffusion, the mean number of particles \( A \) is shown to decay exponentially to a steady state which depends on the details of the initial conditions. The nature of this dependence is demonstrated both analytically and numerically. In contrast, when diffusion is incorporated, it is shown that the mean number of particles \( ⟨N(t)⟩ \) decays asymptotically as \( t^{-d_f/2} \), the memory of the initial conditions being now carried by the dynamical power law exponent. The latter is fully determined by the fractal dimension \( d_f \) of the initial conditions.

Introduction. – In classical approaches to reactive processes mean-field (MF) theories have been useful to predict many non-trivial dynamical and steady state properties such as multistability, periodic concentration oscillations, chaotic motion etc. As long as fluctuations in concentration and occupation number space remain small, MF approaches provide a suitable description of the process. This is the case in many practical situations, where inhomogeneous fluctuations can be efficiently removed from the system by continuous external stirring of the reactants or sufficiently fast internal diffusion.

On the other hand, considerable interest has been devoted to reactive processes taking place on low-dimensional supports in recent years. In such systems, external stirring may prove difficult, and the internal diffusional mixing may not be sufficiently fast to compensate the effect of the restricted support geometry. Correlated spatial fluctuations initially present in the system or induced by the interplay between the chemistry and the spatial characteristics of the support may then give rise to anomalous temporal behaviour and even deviations from the MF steady state. In particular, the outcome of the reaction may be strongly influenced by the dimensionality of the support.

In recent years, the important role of dimensionality was recognized to be an issue common to many statistical processes as well. Fractal supports, which can be produced to display

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a continuum of dimensionalities, have been used widely in experimental \[6,7\] and theoretical studies \[8\] to illustrate the new effects arising in the above context. A second factor of importance in the interplay between local dynamics and transport processes is the form of the initial conditions \[9\]. Depending on whether they are homogeneous, short-ranged or correlated on a long range they may favour quick mixing by diffusion, delays in such a mixing resulting in segregation or, in the limit of immobile reactants, nonergodic behaviour altogether. The objective of the present work is to analyze the evolution of a low dimensional system subject to initial conditions corresponding to a fractal spatial distribution of reactants in the lattice. In particular, the role of the fractal dimension \(d_f\) of the initial distribution will be assessed.

The specific system we shall analyze here is the reaction \(A + A \rightarrow A\) \[10,11\]. The interest in the dynamics of this reaction on fractal supports first arose in connection with exciton fusion experiments on fractal percolation clusters \[6,7\]. While previous work has treated the case of random homogeneous initial conditions \[12\] and the general inhomogeneous case \[13\], we focus here on the specific case of an inhomogeneous fractal distribution. We consider the cases where a) the reactants \(A\) are immobile on a fractal subset of a 1D lattice and b) the reactants \(A\) are initially placed on the fractal subset, but for \(t > 0\) they can diffuse and react throughout the entire 1D lattice. We show, analytically and numerically, that for immobile reactants the steady state reactant concentration does not depend only on \(d_f\) but also on the details of the fractal subset. In contrast, for diffusing reactants we show that the memory of the initial fractal distribution is carried by the anomalous dynamical exponent which is fully determined by \(d_f\).

**Dynamics on fractals: immobile reactants.** – To investigate the effect of fractal initial conditions on reactive systems with immobile reactants we consider the reaction \(A + A \rightarrow A\) on two different Cantor-like sets \(C_1\) and \(C_2\). Set \(C_1\) is obtained by iteration of the segment 1110, while set \(C_2\) is formed by iteration of the segment 1101. After the \(n\)-th iteration, the lattice size \(L\) defined for both sets contains \(4^n\) sites. In both sets, the number of ones and zeros is \(3^n\) and \(4^n - 3^n\) respectively. We consider the ”1” sites as the active sites (sites where reactions can take place) and the ”0” sites as inactive, or empty sites. Both sets are deterministic fractals with \(d_f = \log(3)/\log(4)\). For convenience, we shall number the lattice sites from 1 to \(L\), say from left to right, regardless of whether they are ”0” (inactive) or ”1” (active) sites.

Next, we fill the ”1” sites of set \(C_1\) or set \(C_2\) with particles \(A\) and let the reactions proceed according to the following Monte Carlo (MC) algorithm: at each time step \(\Delta t\), a lattice site \(i\) is randomly chosen. If \(i\) and a randomly chosen neighbour \(i \pm 1\) are occupied, the particle \(A\) at site \(i\) is removed from the lattice with probability \(k_R\). In our model, the site choice is unbiased, i.e. sites \(i + 1\) and \(i - 1\) are chosen with equal probability. On the other hand, \(k_R\) is the conditional probability of reaction at each time step given that two neighbouring sites are occupied and plays thus the role of a local reaction rate. We choose fixed boundary conditions by introducing two additional sites 0 and \(L + 1\) at the boundaries and specifying them as ”0” sites. This particular choice of boundary conditions is selected for convenience and does not play any important role in the large \(L\) limit. Finally, we set \(\Delta t = \frac{1}{L}\) \((1)\),

We are interested in the time evolution of the mean particle number \(\langle N(t) \rangle\) and the associated mean concentration \(\theta(t) := \langle N(t) \rangle / L\). Clearly, with the above choice of initial conditions, \(\langle N(0) \rangle\) diverges while \(\theta(0)\) vanishes in the limit \(L \rightarrow \infty\) as \(L^{(\log(3)/\log(4))^{-1}} = L^{d_f - 1}\). In order to obtain a well posed problem, we shall therefore consider the case of a finite system, as it is the case in experimental situations involving, for instance, mesoscopic scale devices (micelles, single crystallographic faces of a solid catalyst, etc.). Our objective will be to see whether some generic trends will nevertheless show up for long times and/or

\((1)\) This choice implies that all lattice sites are scanned once on average after one time unit.
large sizes \( L \). We expect that the final number of particles \( \langle N(\infty) \rangle \) will strongly depend on the specific form of the initial condition due to the lack of mixing, since particle islands evolve independently from each other. Thus, the spatial correlations present in the initial distribution will propagate in time.

The function \( \langle N(t) \rangle \) can be obtained heuristically by means of the following observation: a string of \( k \) consecutive sites (\( k \)-tuplet) can be destroyed either by internal reaction events between particles inside the tuplet or by reaction between the particles at each edge site of the tuplet and particles sitting at occupied nearest neighbour sites outside the \( k \)-tuplet. The latter events require the existence of a \( k+1 \)-tuplet. The dynamics of the mean number of \( k \)-tuplets \( M_k^{(n)}(\tau) \) (averaged over an ensemble of identical lattices) in a fractal set of size \( L = 4^n \) is given by the following hierarchy of equations \[12, 13\]:

\[
\frac{d}{d\tau} M_k^{(n)}(\tau) = -(k - 1) M_k^{(n)}(\tau) - M_{k+1}^{(n)}(\tau), \quad k = 1 \cdots k_{\text{max}},
\]

where \( k_{\text{max}} \) is the size of the largest \( k \)-tuplet and \( M_k^{(n)}(\tau) \equiv 0 \) for \( k > k_{\text{max}} \). The reaction rate \( k_p \) has been absorbed in the adimensional time variable \( \tau = k_R t \). The general solution of eqs. \[11\] depends strongly on the details of the fractal set. It reads

\[
M_k^{(n)}(\tau) = e^{-(k-1)\tau} \sum_{s=0}^{k_{\text{max}}-k} \frac{(e^{-\tau} - 1)^s}{s!} M_{k+s}^{(n)}(0).
\]

For sets \( C_1 \) and \( C_2 \), \( k_{\text{max}} = 3 \). The mean number of particles (singlets) is then

\[
\langle N(\tau) \rangle = M_1^{(n)}(\tau) = \sum_{s=0}^{2} \frac{(-1)^s}{s!} M_{1+s}^{(n)}(0) + (M_2^{(n)}(0) - M_3^{(n)}(0)) e^{-\tau} + \frac{M_3^{(n)}(0)}{2} e^{-2\tau}.
\]

In particular, the initial tuplet distribution for set \( C_1 \) is given by the number of particle islands \( = 3^{n-1} \) times the number of \( k \)-tuplets contained by each island. In this case, there are three singlets, two doublets and one triplet in each island, \( i.e. \ M_1^{(n)}(0) = 3 \cdot 3^{n-1} = 3^n, \ M_2^{(n)}(0) = 2 \cdot 3^{n-1} \) and \( M_3^{(n)}(0) = 1 \cdot 3^{n-1} \). Asymptotically, we have

\[
\langle N(\infty) \rangle = \sum_{s=0}^{2} \frac{(-1)^s}{s!} M_{1+s}^{(n)}(0) = \frac{3^n}{2} = \frac{\langle N(0) \rangle}{2} = \frac{3}{2} \cdot 3^{n-1}.
\]

This gives a survival factor \( \eta(n) := \langle N(\infty) \rangle / \langle N(0) \rangle = \frac{1}{2} \), \( i.e. \) the number of particles drops to half the initial value regardless of the lattice size. As emphasized by eq. \[11\], each island yields asymptotically a mean number of particles equal to \( \frac{3}{2} \). In contrast, a simple-minded combinatorial counting giving each final state of the island \( (\text{ASA}), (\text{SAS}), (\text{SSA}), (\text{ASS}) \) the same statistical weight yields the wrong factor \( \frac{5}{2} \). This reflects the nonergodicity of the system, implying that the number of statistical paths leading to each steady state is different.

For set \( C_2 \), the initial distribution is slightly more complex and contains a variety of island sizes. We have \( M_1^{(n)}(0) = 3^n, \ M_2^{(n)}(0) = \frac{1}{2}(3^n - 1) \) and \( M_3^{(n)}(0) = \frac{1}{2}(3^{n-1} - 1) \). Using again eq. \[12\], we obtain \( \langle N(\infty) \rangle = \frac{7}{12} 3^n + \frac{1}{2} \) and \( \eta(n) = \frac{7}{12} + \frac{3^n}{4} \). In the large \( L \) limit, \( \eta(n) \rightarrow \frac{7}{12} \approx 0.583 \). The size distribution of the islands plays a crucial role to determine the number of surviving particles. Particles in smaller islands have a higher survival expectancy. Therefore, more particles survive in set \( C_2 \) than in set \( C_1 \).

For set \( C_2 \), the combinatorial argument is again based on the size distribution of the islands, which is connected with the tuplet distribution through the equation \( I_k^{(n)}(\tau) = M_k^{(n)}(\tau) - \)
We then have \( I_{1}^{(n)}(0) = I_{2}^{(n)}(0) = \frac{1}{2}(3^{n-1} + 1) \) and \( I_{3}^{(n)}(0) = \frac{1}{2}(3^{n-1} - 1) \). According to the combinatorial counting, islands of size 1 and 2 will yield one particle asymptotically, whereas island of size 3 yield \( \frac{3}{2} \) particles on average. This is again wrong since, as we know, each three-particle island is reduced to \( \frac{3}{2} \) particles on average.

The time evolution of \( \langle N(t) \rangle \) is easily computed for both fractal sets by substituting the corresponding expressions for the \( k \)-tuplet distributions into eq. (3). In the long time limit, the dominant term describing the decay to the steady state is proportional to \( e^{-\tau} \) (recall that \( \tau = k f R t \)). Thus, the information on the initial distribution is contained in the coefficient of the dominant term rather than in the relaxation time \( k^{-1} \) given by the exponent of the dominant term. In particular, this means that fractals with different \( d_f \) may relax at the same speed into the steady state. We therefore conclude that in the immobile reactant case neither the dynamics nor the steady state are suitably characterized by \( d_f \).

To confirm our results, we have performed MC simulations over \( 2 \cdot 10^3 \) statistical runs on a lattice with \( L = 4^5 = 1024 \) sites. For both fractal sets, the asymptotic concentrations \( \theta(\infty) \) and the dynamics dictated by eq. (3) agree very well with the simulations (fig. 1).

Let us compare the previous results for inhomogeneous initial conditions with the case of a lattice containing only "1" sites. A lattice of length \( L = 4^n \) can be regarded as an iteration of, say, the segment 1111. The initial \( k \)-tuplet distribution is given by \( M_k^{(n)}(0) = 4^n - k + 1 \) with \( k = 1, \ldots, 4^n \). In the large \( L \) limit, this yields \( \langle N(\infty) \rangle = \sum_{s=0}^{4^n-1} (4^n - s)/s! \approx e^{-1}(4^n - 1) \) and \( \eta(n) \approx e^{-1}(1 - 4^{-n}) \), i.e., \( \eta(n) \to e^{-1} \approx 0.367 \) as \( L^{-1} \). As expected, the homogeneous system is characterized by a lower survival factor than any Cantor-like set for all values of \( L \).

A comment on the new features brought in by the fractal initial conditions analyzed above is now in order. The initial inhomogeneities imposed by sets \( C_1 \) or \( C_2 \) decouple the dynamics of different parts of the system and decompose it into smaller, homogeneous subsystems. In principle, they have the same effect as the reaction-induced inhomogeneities, i.e., lowering the number of active sites. However, the interest of considering inhomogeneous initial conditions lies in the fact that, while the initial spatial correlations range over the whole system size, chemically-induced correlations are short-ranged; the latter may only develop between sites initially belonging to the same island. Due to the absence of diffusion or any other randomizing mechanism, a detailed memory of the initial spatial structure is carried by \( \theta(\tau) \) for all times.

As found above, the system’s memory in this case is not sufficiently characterized by \( d_f \). The mean coordination number \( z \) of the fractal subset, defined as the spatial average of the number of active neighbour sites, is not adequate either for the description the dynamics or the steady state. Indeed, the fractal set obtained by the iteration of the segment 1110011110011000 has the same value of \( d_f \) and \( z = \frac{4}{3} \) as set \( C_1 \), but its dynamics and steady state are not the same due to the different \( k \)-tuplet distribution.

An alternative analytical description for the reaction on linear sets is provided by the theory of Markov chains. We have seen that the reaction dynamics of, say, set \( C_1 \), can be fully determined by knowledge of the evolution of a single three-particle island. An island can never evolve into the empty state (SSS). There are 7 possible states (\( \theta \)) of such an island with at least one particle, namely (AAA), (SAA), (ASA), (AAS), (SSA), (SAS) and (ASS). Let us denote them by 1, 2, 3, 4, 5, 6 and 7 respectively. The state vector of the system at time \( t \) is \( \hat{P}(t) = (P_1(t), \ldots, P_7(t))^T \), where \( P_n(t) \) is the probability that the island be in the state \( n \) at time \( t \). \( \hat{P}(t) \) satisfies the stochastic evolution equation

\[
\hat{P}(t + \Delta t) = W^T \hat{P}(t), \tag{5}
\]

\( (\)The number of relevant states can be decreased by symmetry considerations, but we shall keep all seven states for the sake of clarity.\( )
where the elements $w_{i,j}$ of the transition probability matrix $W$ are computed from the MC algorithm for the reaction by counting the number of paths leading from one state to another:

$$
W = \begin{pmatrix}
1/3 & 1/6 & 1/3 & 1/6 & 0 & 0 & 0 \\
0 & 2/3 & 0 & 0 & 1/6 & 1/6 & 0 \\
0 & 0 & 1 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 2/3 & 0 & 1/6 & 1/6 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 1
\end{pmatrix}
$$  \quad (6)

(we have set $k_R = 1$ for simplicity). This matrix possesses four absorbing states and is therefore non-ergodic \[15\]. The steady states $\Pi$ are the eigenvectors of the matrix $W^T$ corresponding to the degenerate eigenvalue $\lambda_1 = 1$. The algebraic multiplicity of $\lambda_1$ is 4, equal to the dimension of the subspace spanned by the four eigenstates $\Pi_i$ with the components $\Pi_{1,j} = \delta_{3,j}, \Pi_{2,j} = \delta_{5,j}, \Pi_{3,j} = \delta_{6,j}$ and $\Pi_{4,j} = \delta_{7,j}$, where $j = 1, \ldots, 7$. The general form of the steady state $\Pi$ can be expressed as a superposition of the states $\Pi_i$, i.e. $\Pi = \sum_i c_i \Pi_i$ with $\sum_i c_i = 1$. The coefficients $c_i$ depend on the initial conditions and can be calculated numerically by iteration of the evolution equation \[9\]. Thus, if one starts with a full island (state 1), the asymptotic mean number of particles on the island will be $\langle N(\infty) \rangle_{isl} = 2 \cdot \frac{1}{6} + 1 \cdot \frac{1}{3} + 1 \cdot \frac{1}{3} + 1 \cdot \frac{1}{3} = \frac{5}{3}$, i.e. we recover the result obtained previously for the Cantor set $C_1$. $\langle N(\infty) \rangle$ can be computed similarly for the set $C_2$ and for any other Cantor-like sets.

The other eigenvalues of $W^T$ describe the decay of $\langle N(t) \rangle$ to the steady state, i.e. $\langle N(t) \rangle - \langle N(\infty) \rangle = \sum_{i=2}^{3} u_i e^{-\lambda_i t}$ with time-independent coefficients $u_{2,3}$. They are $\lambda_2 = \frac{2}{3}$ (twice) and $\lambda_3 = \frac{1}{3}$. This time dependence is corroborated by eq. \[3\], except that the arguments in the exponential functions differ from our result by a factor of $\frac{1}{3}$. However, this artefact is a direct consequence of the choice for the time unit: in the model for the $k$-tuplets and in the MC simulations, it was the time needed to scan the whole lattice, while here it has been implicitly assumed to be the time required to update a single site in a three-site lattice.

Dynamics on fractals: diffusing reactants. – Reactive events in the above diffusionless systems can be viewed as particle jumps into already occupied sites. One can also allow for additional diffusion events, i.e. particles can jump into the "0" sites of the fractal sets with a probability rate $k_D$. The "0" sites can then no longer be considered as inactive sites but rather as initially empty sites. A particle initially placed on a "1" site of the fractal set can now diffuse into a "0" (empty) neighbour site. The latter becomes then a "1" site (occupied), while the original site becomes "0" (empty). On long time scales, this gives classical diffusion. A description in terms of islands is no longer suitable, since they can now interact with each other by means of diffusing particles at the boundaries of each island.

To study the long-time dynamics, we have performed a series of MC simulations. Initially, particles are put on the "1" sites of the lattice and then they start to diffuse freely over the entire lattice and react with each other. In this case we take reflecting boundary conditions, i.e. when particles arrive at sites 0 or $L + 1$, they bounce off and go back to sites 1 and $L$, respectively. Again, the time scale is set by $L^{-1}$. The reaction rate $k_R$, taken to be equal to the diffusion rate $k_D$, can be again absorbed in the time scale by setting $\tau = k_R t$.

For comparison with the diffusionless case, we have used as initial conditions the fractal sets $C_1$ and $C_2$ and have performed the same number of runs ($2 \cdot 10^3$) for $L = 4^5$. The time evolution of the concentration $\theta(\tau)$ has been monitored for sets $C_1$ and $C_2$. Figure 2 displays a double-logarithmic plot of numerical MC results for $\theta(\tau)$ as a function of $\tau$. The dynamical
exponent is given by the slopes of the point curves, which are represented by dots for $C_1$ and crosses for $C_2$; for both sets, the mean slope is very close to the value $-d_f/2$ represented by the continuous straight line. In contrast to the immobile reactant case, $d_f$ appears to be the natural parameter associated to the long time decay of the concentration.

The above analysis suggests a power law behaviour $\theta(t) = \alpha t^{-d_f/2}$ governing the long time dynamics. This law exhibits two types of memory effects: a memory of the dimensionality $d_f$ of the initial particle distribution entering via the exponent; and a more detailed memory of the initial distribution, entering via the amplitude factor $\alpha$. It is only in the regime of extremely long times $t \gg L^2$ that the decay curves for sets $C_1$ and $C_2$ fall into each other. This limit is to some extent trivial from the standpoint of many-particle dynamics, since only one particle remains on the lattice. In the long time regime, detailed information about lacunae in the initial particle distribution will also be kept before reaching the steady state in higher order quantities like the distribution function for the interparticle distance.

These results agree with recent theoretical predictions. When $k_D = k_R$, a closed analytic description in terms of empty $k$-tuplets (also termed intervals) is possible. This method has been used to show that, for fractal initial conditions with $d_f < 2$, $\langle N(t) \rangle$ behaves as $t^{-d_f/2}$ at long times in the large $L$ limit; for $d_f = 2$, logarithmic corrections are necessary, while MF theory applies for $d_f > 2$. In our case $d_f = \log(3)/\log(4) \approx 0.792$, so the exponent "remembers" the initial particle distribution.

Conclusions. – In the present work the reaction process $A + A \rightarrow A$ with fractal initial conditions has been studied both for immobile and diffusing reactants. In the diffusionless case, it was shown that the fractal dimension $d_f$ does not suffice to characterize the dynamics and the steady state. The number of surviving particles at the steady state depends on the details of the initial distribution. In the presence of diffusion, multistationarity is suppressed and the steady state becomes universal and MF-like. However, a long tail characteristic of anomalous dynamics subsists, implying that the decay is governed by a power law $\langle N(t) \rangle \propto t^{-d}$ rather
than an exponential. In contrast to the immobile reactant case, the memory of the initial condition is carried by the characteristic dynamical exponent \( d \) rather than by the steady state, where \( d \) is fully determined by \( d_f \). As expected, the memory of the initial condition is less detailed than in the immobile reactant case, due to the randomizing effect of diffusion.

The above results may be relevant for a series of experimental situations involving systems other than particle aggregates. A few examples are heterogeneous catalysis, evaporation-deposition systems, porous media, percolation clusters and ferromagnetic systems.

Our work can be generalized in many different ways. One can e.g. consider the case of random rather than deterministic fractal initial conditions. Other possibility is studying more complex reactive schemes like the reversible case \( A + A \leftrightarrow A \) \(^3\), whose long-time dynamics still remains to be characterized in detail in the limit of immobile reactants.

** We thank K. Karamanos, Dr. A. Shabunin, Prof. V. Astakhov, G. A. Tsekouras, H. L. Frisch, F. Vikas and F. Baras for helpful discussions. This work was supported, in part, by the NATO Collaborative Linkage Grant No PST.CLG.977654 and the Interuniversity Attraction Poles program of the Belgian Federal Government.

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