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Reaction spreading on graphs

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We study reaction-diffusion processes on graphs through an extension of the standard reaction-diffusion equation starting from first principles. We focus on reaction spreading, i.e., on the time evolution of the reaction product $M(t)$. At variance with pure diffusive processes, characterized by the spectral dimension d_s, the important quantity for reaction spreading is found to be the connectivity dimension d_l. Numerical data, in agreement with analytical estimates based on the features of n independent random walkers on the graph, show that $M(t) \sim t^{d_l}$. In the case of Erdös-Renyi random graphs, the reaction product is characterized by an exponential growth $M(t) \sim e^{\alpha t}$ with α proportional to $\ln\langle k\rangle$, where $\langle k\rangle$ is the average degree of the graph.

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A huge variety of different problems in chemistry, biology, and physics deal with reactive species in nontrivial substrates [1]. Seminal works on reaction and diffusion dynamics date back to the Fisher-Kolmogorov-Petrovskii-Piskunov (FKPP) model [2]

$$\partial_t \theta = D\Delta\theta + f(\theta)\,, \tag{1}$$

where D is the molecular diffusivity, $f(\theta)$ describes the reaction process, and the scalar field θ represents the fractional concentration of the reaction products. Afterward, reaction-transport dynamics attracted a considerable interest for their relevance in a large number of chemical, biological, and physical systems [1].

Complex networks are a recent branch of graph theory becoming very important to different disciplines ranging from physics to social science, from biology to computer science [3]. An impressive amount of work has been done on the study of both complex networks and reaction-transport processes, but is missing a modelistic approach able to conjugate the two fields starting from first principles.

There are two main approaches to studying reaction dynamics on graphs. One concerns agent-based models (Lagrangian description) in which random walkers move on the graph and interact, with a given reaction rule, when they occupy the same site at the same instant [4]. A different approach is based on a mesoscopic description of the reaction dynamics [Eq. (1)] in which diffusion is modified introducing a proper transport term taking into account the feature of the media in which the dynamic takes place [5]. A particular approach in the mesoscopic description of the dynamics (used, e.g., in the recent field of epidemic spreading [6]) is to use a mean-field approximation in which a renormalized reaction term takes into account the network characteristics.

The goal of this Rapid Communication is to study reaction spreading on graphs by extending model (1). In the presence of more general transport processes, the diffusion term $D\Delta\theta$ in Eq. (1), can be replaced by a suitable linear operator $\hat{L}$. A general evolution equation for θ is

$$\partial_t \theta = \hat{L}\theta + \frac{1}{\tau} f(\theta). \tag{2}$$

where we write explicitly the typical time scale τ of the reaction process. An important class of processes of this type is the advection-reaction-diffusion (ARD) class, where $\hat{L} = -\mathbf{u}\cdot\nabla + D\Delta$. Another interesting case is ruled by the effective diffusion operator $\hat{L} = \frac{1}{r^{d_f-1}}\frac{\partial}{\partial r}((r)r^{d_f-1}\frac{\partial}{\partial r})$ [7] suitable to studying reaction dynamics on fractals [5].

Equation (2) is constituted by two terms: the transport term $\hat{L}\theta$ and the nonlinear local reaction $f(\theta)/\tau$. In the limit case without reaction, the link between the solution $\theta(\mathbf{x},t)$ and a suitable stochastic process is quite clear: for instance, if $f(\theta) = 0$ and $\hat{L} = -\mathbf{u}\cdot\nabla + D\Delta$, Eq. (2) is nothing but the Fokker-Plank equation associated with the Langevin equation $d\mathbf{x}/dt = \mathbf{u} + \sqrt{2D}\boldsymbol{\eta}$. In general, even in the presence of reactive terms and for general $\hat{L}$, it is possible to write $\theta(\mathbf{x},t)$ in terms of trajectories using the Freidlin formula [8]:

$$\theta(\mathbf{x},t) = \left\langle \theta(\mathbf{x},0)\exp\left(\frac{1}{\tau}\int_0^t \frac{f(\theta(\mathbf{x}(s;t),s))}{\theta(\mathbf{x}(s;t),s)}ds\right)\right\rangle, \tag{3}$$

where the average is performed over all the trajectories $\mathbf{x}(s;t)$ starting in $\mathbf{x}(0)$ and ending in $\mathbf{x}(t;t) = \mathbf{x}$. The possibility to write the generalization of Eq. (3) for a generic diffusive process has been discussed in [9]. Following this approach we can determine the dynamical equation of reaction diffusion on graphs.

As a diffusion process, we considered diffusion on an undirected, unweighted, and connected graph $G = (V,E)$, where V is the set of vertices of the graph (we consider a finite number N of vertices) and E is the set of edges connecting the vertices. The graph can be represented by its adjacency matrix A_{ij} given by [10]

$$A_{ij} = \begin{cases} 1 & \text{if} \quad (i,j) \in E, \\ 0 & \text{if} \quad (i,j) \notin E. \end{cases} \tag{4}$$

The discrete Laplacian of the graph Δ_{ij} [10,11] is defined by $\Delta_{ij} = A_{ij} - k_i \delta_{ij}$ where $k_i = \sum_j A_{ij}$, the number of nearest neighbors of i, is the degree of vertex i. Once the rate of the jump process w is introduced, the diffusion term can be written as

$$\frac{d\theta_i}{dt} = w \sum_j \Delta_{ij} \theta_j, \tag{5}$$

where θ_i is the concentration at vertex i. Our goal is to add to this equation a reaction term:

$$\frac{d\theta_i}{dt} = w \sum_j \Delta_{ij} \theta_j + \frac{1}{\tau} f(\theta_i). \tag{6}$$

The discrete-time version of the diffusion equation (5) is nothing but the random walk process described by the master equation

$$\theta_n(t + \Delta t) = \sum_j P_{j\to n}^{(\Delta t)} \theta_j(t), \tag{7}$$

where jumps occur at time $\Delta t, 2\Delta t, \ldots$, and the probability for a walker being at vertex i to jump to the vertex j are given in terms of the adjacency matrix

$$\begin{aligned} P_{i\to j}^{(\Delta t)} &= w A_{ij} \Delta t \qquad \text{if} \quad i \neq j, \\ P_{i\to i}^{(\Delta t)} &= 1 - k_i w \Delta t \quad \text{if} \quad i = j. \end{aligned} \tag{8}$$

The discrete-time version of the reaction term can be defined as a nonzero function only at discrete time when δ-form impulses occur:

$$f(\theta,t) = \sum_{n=-\infty}^{\infty} g(\theta) \delta(t - n\Delta t)\, \Delta t, \tag{9}$$

where $g(\theta)$ is a suitable function. Such a choice for $f(\theta)$ allows a rigorous treatment of the discretization of the reaction term. With the above assumption, for Eq. (6) we have

$$\theta_n(t + \Delta t) = G_{\Delta t} \left(\sum_j P_{j\to n}^{(\Delta t)} \theta_j(t) \right), \tag{10}$$

where $G_{\Delta t}(\theta) = \theta + g(\theta)\Delta t$ is the assigned reaction map. It is worth noting that Eq. (10) can be seen as a numerical method when the integration of Eq. (6) is performed in two steps: diffusion and then reaction [9]. The shape of the reaction map $G_{\Delta t}(\theta)$ depends on the underlying chemical model. For autocatalytic pulled reactions [the FKPP class, where, e.g., $g(\theta) = \theta(1-\theta)/\tau$], characterized by an unstable fixed point in $\theta = 0$ and a stable one in $\theta = 1$ (the scalar field θ represents the fractional concentration of the reaction products; $\theta = 1$ indicates the inert material, $\theta = 0$ the fresh one, and $0 < \theta < 1$ means that fresh materials coexist with products), one can use $G_{\Delta t}(\theta) = \theta + \theta(1-\theta)\Delta t/\tau$. In the following we shall consider this type of reaction.

The most important topological features of a graph can be related to the spectral dimension d_s and the connectivity dimension d_l (also called chemical dimension). The former is related to diffusion processes on graphs and can be defined in terms of the return probability P_{ii} at site i for a random walker by $d_s = \lim_{t\to\infty} -2\frac{\ln P_{ii}(t)}{\ln t}$, or equivalently in terms of the density of eigenvalues of the Laplacian operator [11]. The connectivity dimension measures the average number of vertices connected to a vertex in at most l links, as $N(l) \sim l^{d_l}$. For graphs embedded in a Euclidean space, the fractal dimension d_f [12] should also be considered, describing the scaling of the number of vertices in a sphere of radius r in the Euclidean space, as $N(r) \sim r^{d_f}$. The connectivity and fractal dimension can be different and they are related via the mapping between the two distances r and l [13].

As a typical example of an undirected, unweighted, and connected graph, we show in Fig. 1 the reaction spreading in the T graph [14]. The field θ is initialized to zero in each vertex except the central one in which $\theta_i(0) = 1$. Using Eq. (6) we study the time evolution of the system. An interesting observable for characterizing the spreading of the reaction is the percentage of the total quantity of the reaction product, i.e., $M(t) = \frac{1}{N}\sum_{i\in V} \theta_i(t)$ where N is the total number of vertices. As clearly shown in Fig. 1, $M(t)$ grows as a power law that can be interpreted as follows. Starting from a single vertex with $\theta_i(0) = 1$, after t steps the number of vertices reached by the field is $N(t) \sim t^{d_l}$. Therefore, in the limit of very fast reaction, when each vertex reached by the field is immediately burned (i.e, $\theta_j \to 1$), we can expect

$$M(t) \sim t^{d_l}. \tag{11}$$

Figure 1 confirms that the connectivity dimension is the relevant quantity for reaction spreading on the graph. This behavior can be also understood by thinking of the asymptotic behavior of the reaction process as determined by the spreading of the front in the topological metric of the graph. In this case the characteristic time of reaction, τ, appears only in the prefactor of the exponential.

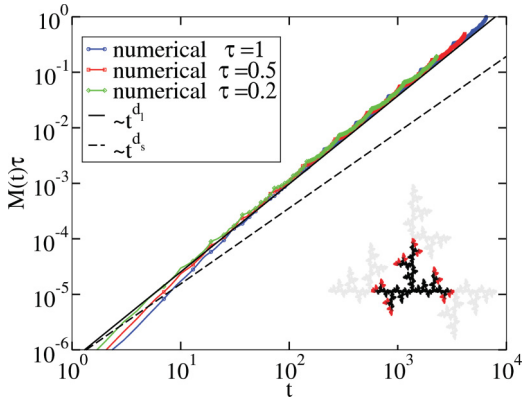

FIG. 1. (Color online) Percentage of quantity of product times τ, $M(t)\tau$ vs t. Numerical results for Eq. (6) with $w = 0.5$ are compared to prediction t^{d_l}. For this graph $d_l = \ln 3/\ln 2 \simeq 1.585$, $d_l = 2\ln 3/\ln 5 \simeq 1.365$. Inset: Spreading on a T fractal.

Moreover, a theoretical argument further confirms the importance of d_l. The analysis is based on an analogy between reaction spreading and the short time regime of the number of distinct sites visited by n independent random walkers after t steps on a graph, $S_n(t)$ [15]. This quantity can be computed as $S_n(t) = \sum_{j=0}^{N} 1 - C_{0j}(t)^n$, where $C_{0j}(t)$ is the probability that a walker starting from site 0 has not visited site j at time t, the sum is over all the N sites of the graph dropping the dependence on the starting site 0. When the number of walkers is large ($n \to \infty$), $C_{0j}(t)^n$ tends to zero if site j has a nonzero probability of being reached in t steps. In this limit, $S_n(t)$ represents all the sites which have nonzero probability of being visited by step t and, because t is equal to the connectivity distance, $S_n(t) \sim t^{d_l}$. This is precisely the regime observed in the reaction spreading [see Eq. (11) and Fig. 1]. An estimate of the validity of the short time regime is given in terms of the smallest nonzero occupation probability on the graph at time t, $P_m = \langle k \rangle^{-t}$, with $\langle k \rangle$ the average degree of the graph, i.e., the mean number of links for each vertex. Because the short time regime is supposed to hold as long as $nP_m \gg 1$, one obtains that the reaction spreading regime is observed up to times $\bar{t} \sim \ln n$. On the other hand, the asymptotic regime is dominated by the number of distinct visited sites by a walker, that is, $S_n(t) \sim t^{d_s/2}$ in graphs with compact exploration $d_s < 2$, or simply by t on graphs with $d_s > 2$ [15]. In the case of a very fast reaction regime, the front can be considered as equivalent to an infinite number of walkers, hence the asymptotic regime is never reached, leaving the dynamics to be governed by the sole d_l.

As for the spectral dimension, it is the relevant quantity when dealing with random-walk dynamics [4] and in some reaction diffusion processes. For instance, in [16] Eq. (6) has been studied for coarsening processes where $f(\theta)$, at variance with our case, has a bistable structure. Moreover a further argument confirms the minor role of the spectral dimension in the case of reaction-diffusion dynamics using FKPP reaction terms. When dealing with standard diffusion [$\langle x^2(t) \rangle \sim t$] it is possible to show [9] that the spreading dynamics is the same displayed by the standard reaction-diffusion problem (1), i.e., $M(t) \sim t^d$ (where d is the dimension of the space). On the other hand, the presence of anomalous diffusion [$\langle x^2(t) \rangle \sim t^{2\nu}$ with $\nu \neq 1/2$] does not imply that the spreading is anomalous: a case exists [9] in which diffusion is anomalous but reaction spreading is standard.

The same behavior displayed in Fig 1 has been observed in several other self-similar graphs (e.g., the Vicsek and Sierpinski carpet, not shown here), confirming the leading role of d_l. In the case of percolation clusters, the importance of the connectivity dimension and the difference between connectivity dimension and fractal dimension were previously shown [13].

Now we focus on the behavior of Eq. (6) for Erdös-Renyi (ER) random graphs [10] for which $d_l = \infty$. In the ER graphs two vertices are connected with probability p. We choose $p > \frac{\ln(N)}{N}$ so that the graph contains a global connected component. The average degree of the graph is $\langle k \rangle = p(N-1)$. On ER graphs the number of points in a sphere of radius l grows exponentially, $N(t) \sim e^{ct}$, hence we expect a similar behavior for the spreading process:

$$M(t) \sim e^{\alpha t}, \tag{12}$$

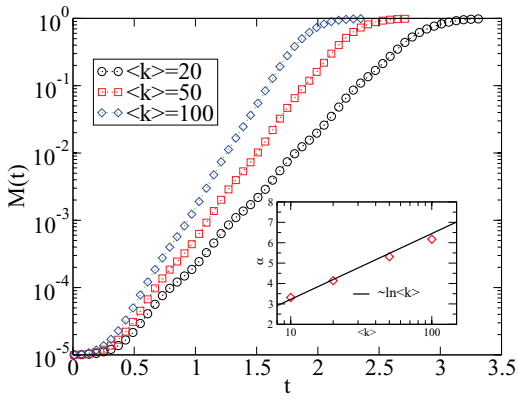

FIG. 2. (Color online) Reaction product $M(t)$ vs t for Erdös-Renyi graphs. Three results for different average degree of connectivity are shown in the fast reaction regime ($\tau = 0.1$). The reaction spreading follows an exponential behavior $M(t) \sim t^{\alpha}$, where α depends on $\langle k \rangle$ as shown in the inset.

as shown in Fig. 2. If $\langle k \rangle$ is large and the reaction is slow enough we have a two-step mechanism: first there is a rapid diffusion on the whole graph, then the reaction induces an increase of θ_i. This leads to a simple mean-field reaction dynamics, $\partial_t \rho(t) = \rho(t)[1 - \rho(t)]/\tau$, where ρ is the average value of θ_i on the graph. In this case $\alpha = 1/\tau$ as clearly observed in numerical simulations (not shown here).

In the much more interesting case of a fast reaction, at each time step the number of sites invaded is proportional to the average degree of the graph, so that after t steps we have

$$M(t) \sim (C_1 \langle k \rangle)^t = e^{C_2 \ln\langle k \rangle t}, \tag{13}$$

leading to $\alpha \sim \ln\langle k \rangle$, see inset of Fig. 2.

Furthermore, at variance with the case of graphs with finite d_l, at least in the case of fast reaction and FKPP reaction term, τ plays an important role since C_2 is a function of τ. Figure 3 shows the dependence of the exponential behavior of reaction spreading rescaled with $\ln\langle k \rangle$ as a function of τ. We can fit the dependence of the curve on τ with $\alpha(k,\tau) \simeq C\tau^{\beta} \ln\langle k \rangle$, with $\beta \simeq -0.8$. This scaling can be related to a mean-field-like

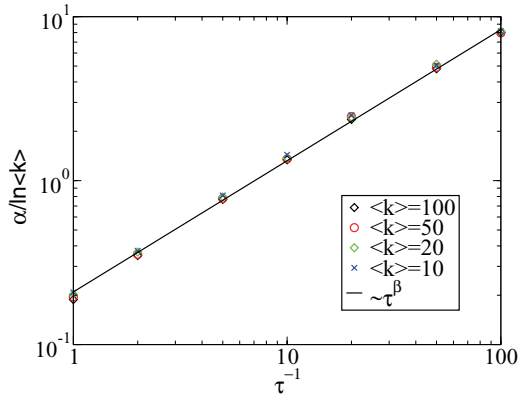

FIG. 3. (Color online) Scaling exponent α normalized with $\ln\langle k \rangle$, as a function of the inverse of reaction time $\frac{1}{\tau}$. The straight line indicates τ^{β} with $\beta = -0.8$.

equation of the type

$$\partial_t \rho(t) = C\tau^{\beta} \ln(\langle k \rangle)\rho(t)[1 - \rho(t)]. \quad (14)$$

We have considered a general model for reaction-diffusion dynamics on graphs, allowing a general and detailed treatment of the diffusive and reaction terms. We study only large systems in which the asymptotic scaling for the reaction spreading is well defined. On the other hand, although the spreading dynamics on small systems is certainly a very interesting issue, it deserves careful attention and it is beyond the scope of the present work. In fact, even in the absence of reaction (i.e., pure diffusion) in small systems the boundaries can induce rather complicated behaviors [17].

On undirected and finite dimensional graphs, we found that a major role in reaction spreading is played by the connectivity dimension, which rules the asymptotic of the reaction product as a function of time. On random graphs with infinite connectivity dimension, reaction spreading shows an exponential behavior, whose scaling depends on the average degree of the graph. In this case, we obtain two mean-field-like equations, one in the slow reaction limit and one in the fast reaction limit. In particular, in the fast reaction case, nontrivial dependence on both the average degree of the graph and the reaction characteristic time is shown. Our approach could be therefore suitable for a rigorous derivation of mean-field-like equations in more complex topologies.

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