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Anomalous Kinetics in A + B → C with Initially-Separated Reactants

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Abstract – We review recent developments in the study of the diffusion reaction system of the type $A+B \to C$ in which the reactants are initially separated. We consider the case where the A and B particles are initially placed uniformly in Euclidean space at $x > 0$ and $x < 0$ respectively. We find that whereas for $d \geq 2$ a single scaling exponent characterizes the width of the reaction zone, a multiscaling approach is needed to describe the one-dimensional system. We also present analytical and numerical results for the reaction rate on fractals and percolation systems.

1 Introduction

The dynamics of diffusion controlled reactions of the type $A + B \to C$ has been studied extensively since the pioneering work of Smoluchowski [1,2]. Most studies have focused on homogeneous systems, i.e., when both reactants are initially uniformly mixed in a d-dimensional space, and interesting theoretical and numerical results have been obtained. When the concentrations of the A and B reactants are initially equal, i.e., $c_A(0) = c_B(0) = c(0)$, the concentration of both species is found to decay with time as, $c(t) \sim t^{-d/4}$ for Euclidean $d \leq 4$-dimensional systems [3-10] and as $c(t) \sim t^{-d_s/4}$ for fractals [5,6] with fracton dimension $d_s \leq 2$. Also, self-segregated regions of A and B in low dimensions ($d \leq 3$) [4] and in fractals [9] have been found. Quantities such as the distributions of domain sizes of segregated regions and interparticle distances between species of the same type and different types have been calculated [11–13]. These systems were also studied theoretically and numerically under steady state conditions and several predictions have been obtained [14–17]. However, the above numerical and theoretical predictions have not been observed in experiments, in part because of difficulties to implement the initially uniformly-mixed distributions of reactants.

In recent years it was realized that diffusion reaction systems in which the reactants are initially separated [18], can be studied experimentally [19,20] and that the dynamics of such a system have several surprising features [20–27]. These systems are characterized by the presence of a dynamical interface or a front that separates the reactants. Such a reaction front appears in many biological, chemical and physical processes [28–34].

Gàlfi and Ràcz [18] studied diffusion-controlled reactions with initially separated reactants. They studied the kinetics of the reaction diffusion process by a set of mean-field (MF) type equations,

$$\frac{\partial c_A}{\partial t} = D_A \frac{\partial^2 c_A}{\partial x^2} - k c_A c_B$$

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$$\frac{\partial c_B}{\partial t} = D_B \frac{\partial^2 c_B}{\partial x^2} - k c_A c_B, \tag{1}$$

where $c_A \equiv c_A(x,t)$ and $c_B \equiv c_B(x,t)$ are the concentrations of A and B particles at position x at time t respectively, $D_{A,B}$ are the diffusion constants and k is the reaction constant. The rate of production of the C-particles at site x and time t, which we call the reaction-front profile, is given by $R(x,t) \equiv k c_A c_B$. The initial conditions are that the A species are uniformly distributed on the right-hand side of $x = 0$ and the B species are uniformly distributed on the left-hand side.

Using scaling arguments Gàlfi and Ràcz [18] find that the width w of the reaction front $R(x,t)$ scales with time as, $w \sim t^{\alpha}$ with $\alpha = 1/6$ and the reaction rate at the center of the front, which is called the reaction height, scales as $h \sim t^{-\beta}$ with $\beta = 2/3$.

Experiments [19] and simulations [19,21–24] for $d \geq 2$ systems in which both reactants diffuse, support the above predicted values for α and β. Indeed, Cornell et al [23] argue that the upper critical dimension is $d = 2$ and the MF approach should therefore be valid for $d \geq 2$. However, numerical simulations of 1D systems show that the width exponent appears to be $\alpha \simeq 0.3$ and the height exponent $\beta \simeq 0.8$ [23,24]. Recently [25] it was argued that α varies between 1/4 and 3/8, depending on the moment at which the width is calculated. For a more detailed discussion, see Sec. III. The origin of the difference between the exponents of 1D systems and those of higher dimensional systems is due to fluctuations in the location of the front which are important in low dimensions and are neglected in the MF approach.

Taitelbaum et al [20,22] studied analytically Eqs. (1) and presented experiments for the limit of small reaction constant or short time. The main results are that several measurable quantities undergo interesting crossovers. For example, the global reaction rate changes from $t^{1/2}$ in the short time limit to $t^{-1/2}$ at the assymptotic time regime. The center of the front can change its direction of motion as found in experiments [20]. Ben-Naim and Redner [26] studied the solution of (1) under steady-state conditions. Generalization of Eqs. (1), by including persistence effects, were studied by Vilensky et al. [35].

2 The Form of the Reaction Front, $R(x,t)$, in the Mean-Field Approach

In a recent work, Larralde et al. [27] considered the symmetric case in which both diffusion constants and concentrations are equal, i.e., $D_A = D_B \equiv D$ and $c_A(x,0) = c_B(x,0) = c_0$. If we define $F(x,t) \equiv c_A(x,t) - c_B(x,t)$, then from Eq. (1) follows,

$$\frac{\partial F}{\partial t} = D \frac{\partial^2 F}{\partial x^2}, \tag{2}$$

subject to the conditions that initially the A particles are uniformly distributed to the right of the origin while the B particles are uniformly distributed to the left of the origin. Equation (2) has the solution $F(x,t) = c_0 \operatorname{erf}(x/\sqrt{4Dt})$.

We rewrite the concentrations of A and B particles as (see Fig. 1),

$$c_A(x,t) = G_1(x,t) + \delta c_1(x,t), \qquad c_B(x,t) = G_2(x,t) + \delta c_2(x,t), \tag{3}$$

where

$$G_1(x,t) = \begin{cases} F(x,t) & [x > 0] \\ 0 & [x < 0], \end{cases} \tag{4}$$

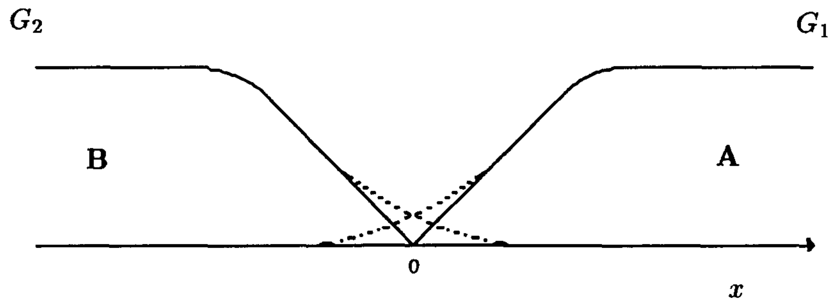

Fig. 1. Schematic picture of the reactant concentration profiles near the origin. The solid lines represent the $G_{1,2}(x,t)$ part of the profile, the dashed lines represent the complete form $G_{1,2}(x,t) \pm \delta c(x,t)$. Note that the profile of species A is given solely by $\delta c(x,t)$ on the left of the origin. (From Ref. [27].)

and $G_2(x,t) = G_1(-x,t)$. It is easy to see that under the above conditions, $\delta c_1(x,t) = \delta c_2(x,t) \equiv \delta c(x,t)$. Substituting Eqs. (3) into Eqs. (1) yields

$$\frac{\partial(\delta c)}{\partial t} = D\frac{\partial^2(\delta c)}{\partial x^2} - k\left(c_0 \operatorname{erf}\left(\frac{x}{\sqrt{4Dt}}\right) + \delta c\right)\delta c. \tag{5}$$

The asymptotic solution for this equation that vanishes as $x \to \infty$ is,

$$\delta c(x,t) \sim t^{-1/3}\left(\frac{x}{t^{1/6}}\right)^{-1/4} \exp\left[-\frac{2}{3}\left(\frac{\lambda x}{t^{1/6}}\right)^{3/2}\right], \quad t^{1/6} \ll x \ll (4Dt)^{1/2}, \tag{6}$$

where $\lambda = (ka/D)^{1/3}$, $a \equiv c_0/(\pi D)^{1/2}$. As may be confirmed by direct substitution, this expression is a solution of Eq. (5) up to terms of order $(\delta c)/t$, which can be neglected for large t.

Using Eq. (6) we can write an expression for the reaction-front profile $R(x,t)$ defined in (1) as,

$$R(x,t) \simeq \frac{kax}{t^{1/2}}(\delta c) \sim t^{-2/3}\left(\frac{x}{t^{1/6}}\right)^{3/4} \exp\left[-\frac{2}{3}\left(\frac{\lambda x}{t^{1/6}}\right)^{3/2}\right]. \tag{7}$$

It is seen that the width of the reaction front grows as $t^{1/6}$, whereas the height can be identified with the prefactor $t^{-2/3}$ in Eq. (7), consistent with the exponents found by Gàlfi and Ràcz [18]. Equation (7) provides a more quantitative solution of Eqs. (1) than the previous scaling arguments [18], as well as information on the dependence of the form of the reaction front on the parameters c_0, k, and D, for the symmetric case.

For the case in which one reactant is static no analytical solution (of Eq. 1) exists for the form of the reaction front. However, numerical solutions of Eq. (1) with $D_B = 0$ shown in Fig. 2, suggest that $R(x,t) \sim t^{-\beta}g(x/t^{\alpha})\exp(-|x|/t^{\alpha})$. The excellent scaling in Fig. 2b suggests that the width does not increase with time, i.e., $w \sim t^{\alpha}$ with $\alpha = 0$ and $h \sim t^{-\beta}$ with $\beta = 1/2$.

3 The Front, R(x,t), in d = 1

The spatial distribution of the C particles in $d = 1$ systems, $R(x,t)$, when both reactants are diffusing with the same diffusion constant, $D_A = D_B \neq 0$, has been calculated numerically [24]. The data shown in Fig. 3, in a scaling form, suggest that

$$c_C(x,t) \equiv \int_0^t R(x,t')dt' \sim \exp(-a|x|/t^{\delta}), \tag{8}$$

with $\delta = 0.37 \pm 0.05$. In a recent work [25] it was found that the reaction front in one dimension is characterized by multiple characteristic length scales. Let $m(t)$ be the midpoint between the rightmost A particle (RMA) and the leftmost B particle (LMB). It is found that $\xi(t) \equiv \langle m^q(t)\rangle^{1/q} \sim t^{\delta}$ with $\delta \approx 3/8$ independent on q. Another length scale that characterizes the reaction front is the distance $\ell_{AB}(t)$ between the RMA and LMB. At time t, a

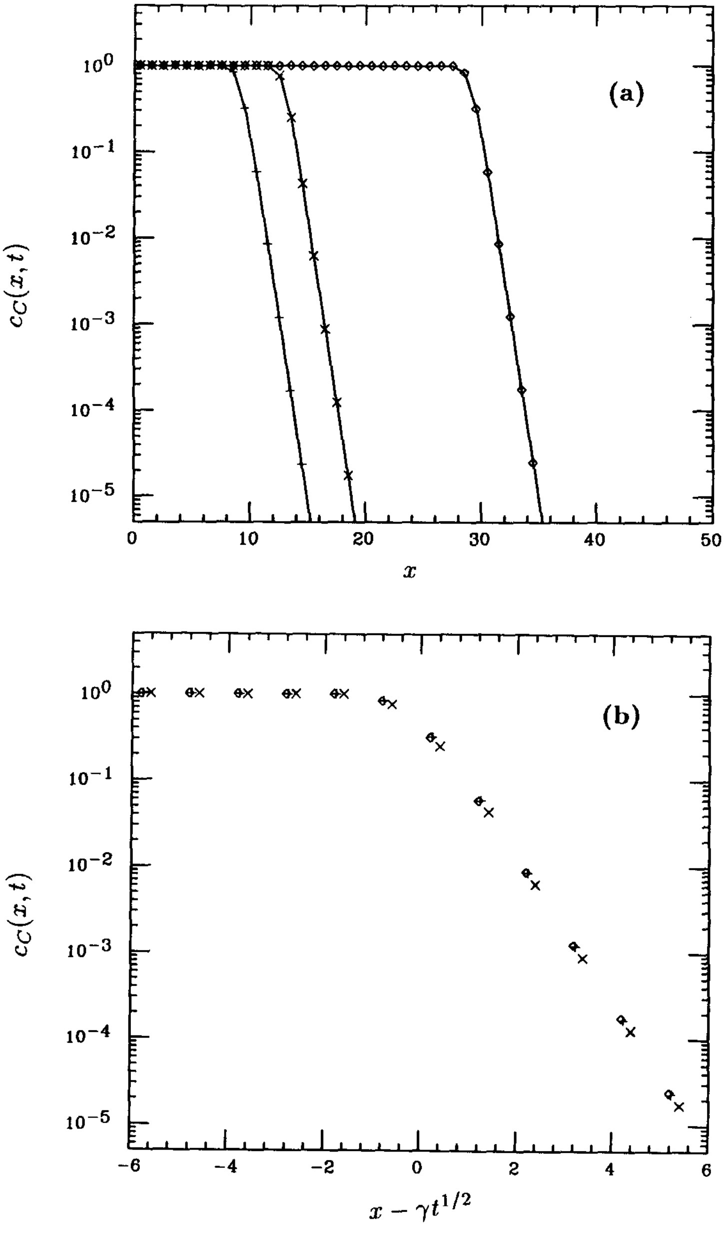

Fig. 2. Numerical solution of Eq. (1) for the case $D_B = 0, D_A \neq 0$: (a) plot of $c(x,t)$ as a function of x for $t = 500, 1000$ and 5000; (b) the good scaling plot of $c(x,t)$ as a function of $(x - \langle x\rangle)$ indicates that $\alpha = 0$ and $\beta = 1/2$.

reaction will occur at position $m(t)$ if and only if $\ell_{AB}(t) = 0$. It is found that $\langle \ell^q_{AB}(t)\rangle^{1/q}$ scales as t^σ with $\sigma \approx 1/4$ for all q. This result is similar to that found by Weiss at al. [36] for the analogous quantity in the one-dimensional trapping problem.

Next, Araujo et al. studied the spatial moments of the reaction front [25],

$$\langle |x|^q \rangle \equiv \frac{\int\limits_0^\infty x^q R(x,t)dx}{\int\limits_0^\infty R(x,t)dx} \sim t^{1/2}\frac{d}{dt}\int\limits_0^\infty x^q c_C(x,t)dx \equiv t^{q\alpha_q}. \tag{9}$$

While both the distributions of $m(t)$ and $l_{AB}(t)$ seem to be describable in terms a of single can scale, the moments of the reaction front do not follow a simple scaling relation. Indeed, the small moments, $q \to 0$, approach the value $\alpha_0 = 1/4$, whereas the high moments approach the value $\alpha_\infty = 3/8$ (see Fig. 4). Note that the value α of the second moment coincides with the value ($\simeq 0.3$) obtained in previous numerical studies [23,24].

To explain the behavior of α_q, we studied numerically the form of $R(x,t)$. Our data suggest that

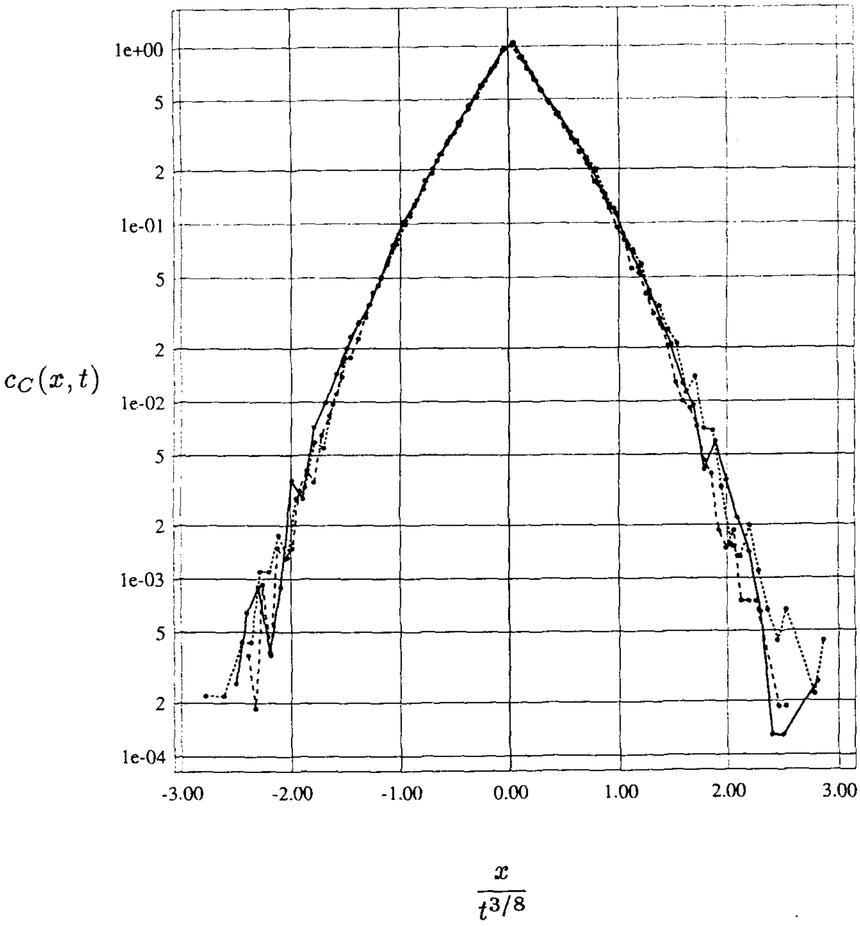

Fig. 3. Plot of $c_c(x,t)$ defined in Eq. (8) for $d = 1$ system with system size $L = 10\,000$ and $t = 1000$, 5000 and 10 000.

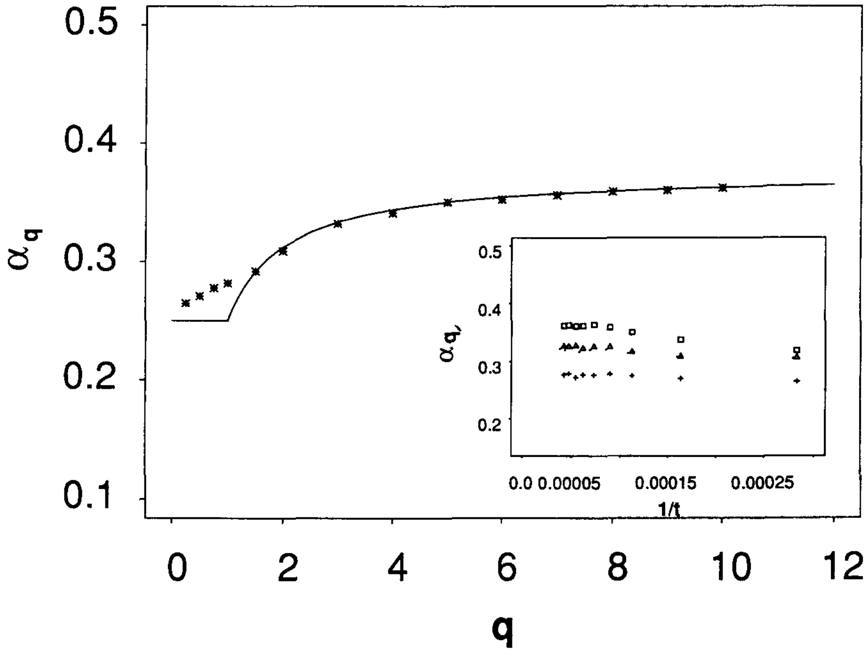

Fig. 4. Plot of α_q as a function of q obtained from extrapolation to infinite times for values of q ranging from 1/4 to 10. The solid line represents the theoretical prediction of Eq. (11). Inset shows successive values of the exponents α_q which characterize the moments $\langle x^q\rangle^{1/q}$ of the reaction as a function of $1/t$ for $q = 1/2(+), 2(\times), 8(\diamond)$.

$$\bar{R}(x,t) \equiv \frac{R(x,t)}{\int_0^\infty R(x,t)dx} \sim u^{-2} e^{-|u|/t^{1/8}}, \tag{10}$$

where $u = x/t^{1/4}$. Substituting this expression into Eq. (9) yields

$$\alpha_q = \begin{cases} 1/4 & q < 1 \\ (3/8) - (1/8q) & q \geq 1 \end{cases} \tag{11}$$

where for $q = 1$

$$\langle |x| \rangle \sim t^{1/4} \ln t. \tag{12}$$

Our results for $q \geq 1$ are in good agreement with these theoretical predictions (see Fig. 4). However for $q < 1$ our results are slightly larger than 1/4, which is probably due to difficulties in reaching the assymptotic limit [37].

For the case $D_A \neq 0, D_B = 0$, analytical and numerical studies [38] yield for the reaction front

$$R(x,t) = \frac{1}{4t^{3/4}} \left(\frac{2\gamma^2}{\mu\pi}\right)^{1/2} \exp\left[-\frac{(x-\gamma t^{1/2})^2}{2\mu t^{1/2}}\right]\left[1 + \frac{x-\gamma t^{1/2}}{2\gamma t^{1/2}}\right], \tag{13}$$

where γ and μ are constants. From Eq. (13) follows that $\alpha = 1/4$ and $\beta = 3/4$. It is interesting to note that the time integral of $R(x,t)$, which is the total production of the C particles at x up to time t is given by,

$$c_C(x,t) = \int_o^t R(x,\tau)d\tau = \frac{1}{2}\mathrm{erfc}\left(\frac{x-\gamma t^{1/2}}{\sqrt{2\mu t^{1/2}}}\right). \tag{14}$$

Larralde et al. [38] performed analogous calculations of $\langle m^q(t)\rangle^{1/q}$, for $\langle \ell_{AB}^q\rangle^{1/q}$, for the case where one of the reactants is static. Numerically and theoretically, results show that in this case the two exponents coincide: $\delta = \sigma = 1/4$.

To summarize, we find four sets of exponents shown in Table 1.

Table 1

	$d=1$	MF $(d>2)$
[both moving]	$1/4 \le \alpha_q \le 3/8$ $\delta = 3/8, \sigma = 1/4$	$\alpha = 1/6$ $\beta = 2/3$
[one static]	$\alpha = 1/4$ $\beta = 3/4$ $\delta = \sigma = 1/4$	$\alpha = 0$ $\beta = 1/2$

4 The Reaction Rate in $d = 2$ Percolation

The case of $A + B \to C$ with initially-separated reactants on fractal systems was studied on the $d = 2$ infinite percolation cluster at criticality [22]. For an illustration of the percolation system see Fig. 5. It is expected that the total number of reactants up to time t,

$$\int_{-\infty}^{\infty} c_C(x,t)dx,$$

scales as the mean displacement of a random walker on a fractal, i.e., $\langle r^2\rangle^{1/2} \sim t^{1/d_w}$, where d_w is the anomalous diffusion exponent [39,40]. From this follows that the reaction rate

$$R(t) \equiv \int_{-\infty}^{\infty} R(x,t)dx \sim t^{-\gamma}, \qquad \gamma = 1 - 1/d_w. \tag{15}$$

One has to distinguish between reaction-diffusion on the infinite cluster and in the percolation system, containing also finite clusters [39]. The reaction rate on the infinite cluster system is smaller and decreases more slowly than that of the system containing clusters of all sizes. This can be understood as follows. At any finite time we can divide all clusters into two groups according to their sizes: *active* clusters—clusters of mass $s > s^*$, in which at time $t < t^* \sim s^{*d_w/d_f}$ particles are not aware of the finiteness of their cluster (this group contains the infinite cluster)—and *inactive* clusters of mass $s < s^*$ on which at least one of the reactants has been vanished and the reaction rate has became zero. According to this picture, in the full percolation system at any time there are active clusters of finite size that can contribute to the reaction rate. Therefore, the rate of reaction in the percolation system is always higher, than on the infinite cluster. Also, at any time there are some finite clusters that become inactive causing additional (comparing with the infinite cluster network) decrease of the rate of reaction in the percolation system. Since the system is self-similar one expects a change in the reaction rate exponent.

To quantify the above considerations we can look on each cluster of mass s and linear size $r \sim s^{1/d_f}$ as a reservoir of particles divided by the *front line* into two regimes: A and B. We introduce an *active front* of a cluster as the sites belonging both to the cluster and to the front line. The length ℓ_s of the *active front* of a single cluster of size s is expected to be

$$\ell_s \sim r^{d_f-1} \sim s^{(d_f-1)/d_f}. \tag{16}$$

Next we assume that the rate of reaction on a cluster of mass s per unit length of active front is

$$R_0(t) \sim \begin{cases} t^{-\gamma} & t < t^* \\ 0 & t > t^* \end{cases}, \tag{17}$$

where $t^* = s^{d_w/d_f}$. Therefore, the total contribution of active clusters of size s to the reaction rate is

$$R_s(t) \sim \varphi_s s^{\frac{d_f-1}{d_f}} t^{-\gamma}, \tag{18}$$

where φ_s is number of clusters of size s that intersect the front line. One can estimate φ_s as follows. In a percolation system of size $L \times L$ there are n_s clusters of mass s. Only a small part of them intersects the front line, those in a strip of width $w \sim s^{1/d_f}$ around the front line. Their fraction is w/L. Therefore, $\varphi_s \sim \frac{s^{1/d_f}}{L} n_s \sim s^{1/d_f} n_s$. Substituting this in (18) we get

$$R_s(t) \sim s^{1/d_f} n_s s^{\frac{d_f-1}{d_f}} t^{-\gamma} = t^{-\gamma} s n_s. \tag{19}$$

Thus, the reaction rate in the percolation system is

$$R(t) = \sum_{s=s^*}^{\infty} R_s(t) = t^{-\gamma}(s^*)^{2-\tau} = t^{-\gamma} t^{-\delta} = t^{-\gamma'},$$

where

$$\delta = \gamma' - \gamma = \frac{d_f}{d_w}(\tau - 2). \tag{20}$$

These results are in agreement with our 2d simulations, shown in Fig. 6.

We also study the finite size effects on the reaction-diffusion system. For a percolation system of size $L \times L$ we expect that for the infinite cluster,

$$R(t) = L^{d_f-1} t^{-\gamma}, \tag{21}$$

while for the percolation system

$$R(t) = L t^{-\gamma'}. \tag{22}$$

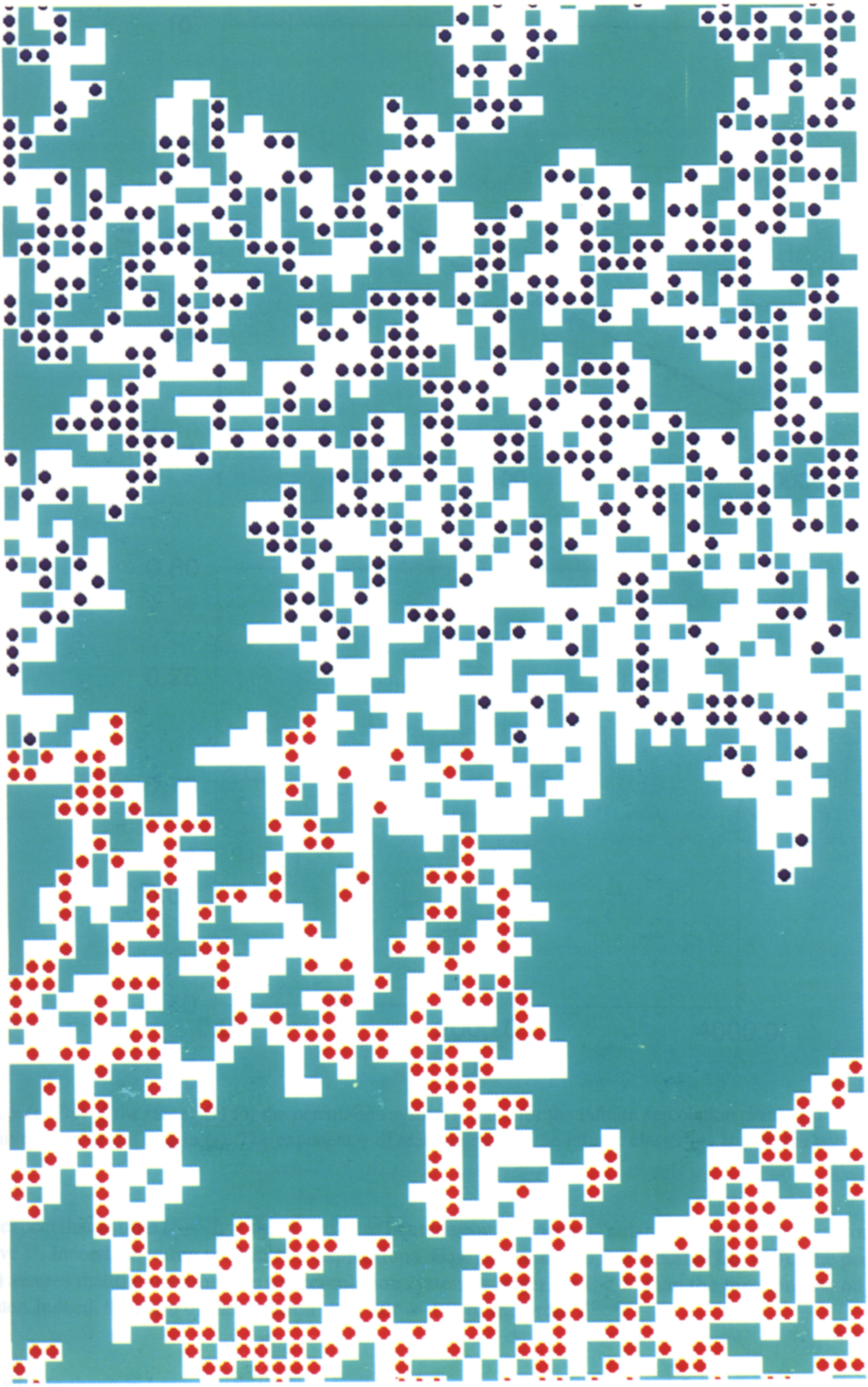

Fig.5. Reaction-difusion on the 2 - dimensional infinite percolation cluster at criticality at $t = 2000$. White squares represent sites of the infinite claster. Blue and red circles represent the A and B particles. Initially all sites of cluster in the right half plane and left half plane were occupied by A and B particles respectively.

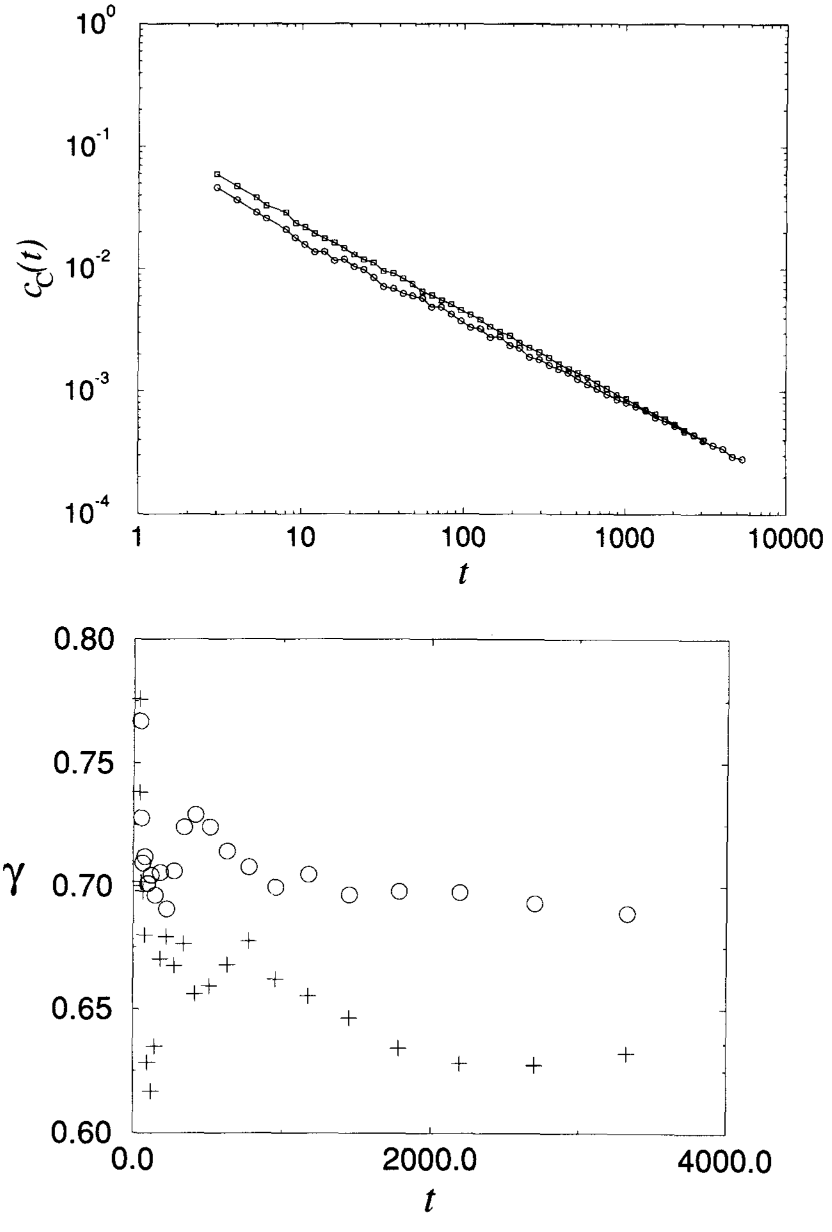

Fig. 6. (a) Plot of the rate $c_C(t)$ for the percolation system (◇) and for the infinite percolation claster (○). (b) Plot of successive slopes of the data in (a). The exponent γ of reaction rate on the infinite claster (+) and on the percolation system (○).

We expect that at time $t^* \sim L^{d_w}$ these two rates become equal, since no "small" active cluster exists in the system above t^*. Indeed, equating the last two expressions, Eqs. (21) and (22), we rederive Eq. (20). The prefactor L in (22) assures that the reaction rate for percolation system is larger for $t < t^*$ than the reaction rate on the infinite cluster. Indeed, the ratio of the reaction rates at $t = 1$ is of the order L^{2-d_f}.

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