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Role of fluctuations for inhomogeneous reaction-diffusion phenomena

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Although fluctuations have been known to change the long-time behavior of homogeneous diffusion-reaction phenomena dramatically in dimensions $d \leq 4$, simulations of reaction fronts in two-dimensional $A+B \rightarrow C$ inhomogeneous systems have only shown marginal departure from mean-field behavior. We perform cellular-automata simulations of the one-dimensional case and find that the width $W(t)$ of the reaction front behaves as $t^{0.293 \pm 0.005}$, in contrast to mean-field behavior $t^{1/6}$. We develop a scaling theory to obtain inequalities for the exponents in the more general mechanism $nA+mB \rightarrow C$. Heuristic arguments about the range of fluctuations imply that the mean-field behavior should be correct in dimensions larger than an upper critical dimension $d_{\mathrm{up}}=2$, irrespective of the values of n and m. This leads us to reinterpret the two-dimensional data obtained previously in terms of a logarithmic correction to mean-field behavior.

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I. INTRODUCTION

Chemical reactions in which several species react are usually described in terms of macroscopic rate equations. These rate equations give the time evolution for the local average concentrations of the different species. They assume that the reaction is completely described by the local average densities, i.e., that the reaction introduces no correlations between the reacting species. This is reminiscent of a mean-field approximation in statistical physics, and therefore neglects an important aspect of the problem, namely, the microscopic fluctuations.

For homogeneous reaction-diffusion systems, it is well known that the fluctuations play an important role. The simplest example may be provided by the annihilation reaction $A+B \rightarrow 0$. The solution of the rate equations predicts that the concentration of A (or B) will decrease at short time as $a \simeq t^{-1}$ while a calculation taking into account the microscopic fluctuations in the particle density a gives $a \simeq t^{-d/4}$, where d is the dimensionality of the system [1].

A more complicated class of problem is that for which the initial condition is inhomogeneous. The inhomogeneities of the problem often arise from the presence of sources or sinks of particles. This may lead to the formation of reaction-diffusion fronts. Those fronts play an important role in many physical or chemical problems. Examples are given by solid-solid chemical reactions [2], catalysis [3], crystal growth [4], diffusion-limited aggregation [5], or precipitate formation [6].

A simple model displaying a reaction front has been analyzed at the level of the rate equations by Gálfi and Rácz [7]. They consider two species A and B diffusing on a two-dimensional substrate and reacting to form a new species C. Provided that the two diffusing reagents A and B are initially separated in space, they will react in a confined region ("reaction front"). An appropriate choice of geometry reduces the problem to one dimension in this approximation. In particular, it is assumed that, at time $t=0$, the system is uniformly filled with A for $x \leq 0$, and uniformly filled with B for $x > 0$. The study of the properties of this front provides relevant information about the production of C particles. The analytic treatment of Gálfi and Rácz assumes that the reaction rate is expressed in terms of the product of mean particle densities, which we shall henceforth refer to as the "mean-field" approximation. Their main result is that the distribution of the production of C particles, i.e., the reaction front, has a scaling form in the large-time limit. In particular, the width of the front behaves as $W(t) \simeq t^{1/6}$.

The role of fluctuations in this system has been recently investigated in two dimensions by numerical simulations of microscopic models [8,9]. While the Monte Carlo simulations of Jiang and Ebner [8] were not precise enough to detect corrections due to fluctuations, the cellular-automata approach of Chopard and Droz [9] showed a significant departure from the value $\frac{1}{6}$ for the width exponent. If the fluctuations play a significant role in two dimensions, their effect in one dimension should be still more dramatic.

In this paper, we present a scaling analysis of the more general process $nA+mB \rightarrow C$, obtaining rigorous inequalities for and relations between the scaling exponents. We then present a thorough analysis of a one-dimensional cellular-automaton model of this reaction-diffusion process in the case $m=n=1$. We find that the reaction width scales like $t^{0.293 \pm 0.002}$, contrasted with the mean-field value $t^{0.1666}$.

Furthermore, we formulate a microscopic argument to obtain the range of fluctuations. We argue that the upper critical dimension, where the fluctuations become of the same range as the mean-field reaction width, is 2, independently of n and m. This leads us to reinterpret the two-dimensional data in terms of a logarithmic correction to the mean-field power law.

In the concluding section, we consider possible schemes for introducing fluctuations at the level of the macroscopic rate equations and suggest further avenues of research.

II. SCALING ANALYSIS

The process we are interested in is the reaction

$$nA+mB\rightarrow C \tag{1}$$

between two species A and B diffusing on a gel, for instance. This reaction-diffusion process can be described by the following partial differential equations:

$$\partial_t a=D\nabla^2 a-k_1 R\,, \quad \partial_t b=D\nabla^2 b-k_2 R\ , \tag{2}$$

where a and b are the local concentrations of A and B, and D the diffusion constant which, for simplicity, is assumed to be identical for both species. If the diffusion constants are different for the two species, we still expect the same asymptotic scaling behavior, with irrelevant corrections to the scaling function [7]. R is the reaction term. It is proportional to the joint probability of having n particles of type A and m particles of type B simultaneously present at the same point. The reaction constants k_1 and k_2 are such that

$$\frac{k_1}{k_2}=\frac{n}{m}\ . \tag{3}$$

This condition ensures that each reaction event consumes n A atoms and m B atoms.

In this work we are interested in a situation where a reaction front develops, as is the case when A and B are initially separated in space. We assume that $a(x,y,\ldots,t=0)=a_0$ if $x<0$ and $a=0$ otherwise. Similarly, $b(x,y,\ldots,t=0)=0$ for $x<0$ and $b=b_0$ for $x>0$. With this choice of initial conditions, Eqs. (2) reduce to a problem containing only the space variable x.

In general, the reaction region (the front) is expected to move through the system as time goes on. Following Gálfi and Rácz [7], one observes that $u\equiv ma-nb$ obeys the diffusion equation

$$\partial_t u(x,t)=D\partial_x^2 u(x,t)\ . \tag{4}$$

With the initial condition $u(x<0,t=0)=ma_0$ and $u(x>0,t=0)=-nb_0$, the solution is found to be

$$u(x,t)=\frac{ma_0-nb_0}{2}-\frac{(ma_0+nb_0)}{\sqrt{\pi}}\int_0^{x/2\sqrt{Dt}}du\,\exp(-u^2)\ . \tag{5}$$

The position of the front $x_f(t)$ can be defined as the point where the rate of reaction is maximal. If $u>0$, $a/n>b/m$ and then the production of C particles is limited by the number of B particles. This number increases as u decreases. Similarly, for $u<0$, the reaction is limited by the number of A particles which increases with u. Therefore $x_f(t)$ is given by $u(x_f(t),t)=0$. From Eq. (5), it is then clear that $x_f(t)$ behaves like $\sqrt{t}$ and that $x_f(t)=0$ if $ma_0=nb_0$. For the sake of simplicity, we shall now consider only the situation where the front is motionless. This does not affect the scaling arguments we present, since the velocity of motion of the front decreases monotonically, and so must asymptotically move more slowly than the particles.

Interesting information on the long-time behavior of a, b, and R can be obtained from Eqs. (2) by using simple physical arguments and assuming the scaling forms

$$\begin{aligned} a(x,t)&=t^{-\gamma_a}\hat{a}(xt^{-\alpha_a})\ ,\\ b(x,t)&=t^{-\gamma_b}\hat{a}(xt^{-\alpha_b})\ ,\\ R(x,t)&=t^{-\beta}\hat{R}(xt^{-\alpha})\ . \end{aligned} \tag{6}$$

These scaling forms are only valid for $x\ll L_d$, where L_d is the width of the depletion zone. Consistency between the forms and Eq. (2) requires

$$\alpha_a=\alpha_b=\alpha,\quad \gamma_a=\gamma_b=\gamma\ . \tag{7}$$

Relations between these exponents can be derived from two assumptions [7,8], based on the fact that A and B react only in a localized region.

(i) The reaction zone increases more slowly than the characteristic length of the diffusion process. This means that, in the long-time limit, a and b vary over a typical length governed by diffusion, i.e., $\mathrm{grad}a\propto\mathrm{grad}b\propto t^{-1/2}$ and thus

$$\gamma+\alpha=\tfrac{1}{2}\ . \tag{8}$$

(ii) Due to the gradient concentration of A and B, a flux of particles towards the reaction region is observed. Assuming that the reaction is fed by these particle currents, the reaction-diffusion equations take on a quasistationary form $D\nabla^2 a=k_1 R$ and $D\nabla^2 b=k_2 R$. For the exponents, this amounts to the relation

$$\gamma+2\alpha=\beta\ . \tag{9}$$

For consistency with assumptions (i) and (ii), we should also have

$$\alpha<\tfrac{1}{2} \tag{10}$$

so that the width of the front grows slower than the depletion zone produced by the diffusion. To ensure the quasistationary of the solution, we need furthermore that

$$\begin{aligned} D\nabla^2 a&\simeq t^{-\gamma-2\alpha}\\ &\gg\partial_t a=-t^{-\gamma-1}(\gamma\hat{a}+\alpha x t^{-\alpha}\hat{a}')\ , \end{aligned} \tag{11}$$

which is again consistent with the condition (10).

From relations (8) and (9) one also gets

$$\beta-\alpha=\tfrac{1}{2}\ , \tag{12}$$

which is easy to check numerically since $\int_{\mathrm{volume}}R\propto t^{-\beta+\alpha}$.

Relations (8), (9), and (12) are still quite general since no explicit form of the reaction term R has been used. In a mean-field approximation, one neglects the fluctuations and assumes that the reaction term factorizes into a product of the local concentrations of A and B. For the process $nA+mB\rightarrow C$, the mean-field reaction rate R_{MF} takes the form

$$R_{\mathrm{MF}}=a^n b^m \ . \tag{13}$$

This introduces another relation among our exponents, namely,

$$\beta=(n+m)\gamma \tag{14}$$

and we obtain the following the values for the mean-field exponents:

$$\alpha_{\mathrm{MF}}=\frac{n+m-1}{2(n+m+1)},\quad \gamma_{\mathrm{MF}}=\frac{1}{n+m+1} \ . \tag{15}$$

Note that these values are compatible with assumptions (i) and (ii) since $\alpha_{\mathrm{MF}}<\frac{1}{2}$ for any finite values of n and m. In addition, a numerical solution of Eq. (2), with $R=R_{\mathrm{MF}}$, have yielded these exponents, for the cases investigated, namely, $(n,m)=(1,1)$, $(n,m)=(2,2)$, and $(n,m)=(3,1)$. When $n=m=1$ this gives the well-known result [7] $\alpha_{\mathrm{MF}}=\frac{1}{6}$ and $\beta_{\mathrm{MF}}=\frac{2}{3}$.

The values of the non-mean-field exponents can be obtained numerically, as discussed in the next section. Although an analytical determination is probably hopeless, an inequality can be derived by considering the ratio

$$\begin{aligned}\frac{R}{R_{\mathrm{MF}}}&=t^{-\beta+(n+m)\gamma}F(xt^{-\alpha})\\&=t^{(n+m-1)/2-\alpha(n+m+1)}F(xt^{-\alpha}) \ ,\end{aligned} \tag{16}$$

where we used relations (8) and (9) and F is a scaling function. With Eq. (15), this ratio also reads

$$\frac{R}{R_{\mathrm{MF}}}\simeq t^{-(n+m+1)(\alpha-\alpha_{\mathrm{MF}})} \ . \tag{17}$$

Since the reaction introduces correlations between the two species which tend to reduce the probability of particles meeting, R/R_{MF} must not diverge when t increases, and we conclude that

$$\alpha>\alpha_{\mathrm{MF}} \ . \tag{18}$$

The quantity $\alpha-\alpha_{\mathrm{MF}}$ is thus a measure of the quality of the mean-field approximation $R\approx a^n b^m$.

III. NUMERICAL SIMULATIONS

The numerical simulations have been made using a cellular-automata (CA) algorithm for the reaction-diffusion of the particles. This algorithm has been used by Chopard and Droz for the two-dimensional case and is described in detail in their paper [9]. Accordingly, we shall simply give the main points for the one-dimensional case.

We assume that the particles are constrained to move on a lattice of spacing λ, either to the left or to the right. At each time step τ, the particles move towards one of their nearest neighbors, according to the direction of their velocity $v_i=e_i\lambda/\tau$, where $e_1=1$ and $e_2=-1$. Diffusion is produced by a simultaneous random walk and reaction takes place at each site where two particles A and B are simultaneously present with opposite velocities. The particles involved in such a reaction process disappear from the lattice and are replaced by a C particle which does not enter furthermore into the dynamics.

We introduce the occupation numbers $a_i(x,t)$ and $b_i(x,t)$ defined as the number of, respectively, A and B particles entering the site x, at time t, with a velocity v_i. In the spirit of a cellular-automata approach, we construct a dynamics such that a_i and b_i can be either 0 or 1 (exclusion principle).

Our CA model is described by the following equations:

$$a_i(x+\lambda e_i,t+\tau)=\sum_{l=0,1}\mu_l(a_{i+l}-k_{i+l}R_{i+l}) \ , \tag{19}$$

$$b_i(x+\lambda e_i,t+\tau)=\sum_{l=0,1}\nu_l(b_{i+l}-k_{i+l+1}R_{i+l+1}) \ , \tag{20}$$

$$R_i=a_i b_{i+1} \ . \tag{21}$$

The right-hand side of Eqs. (19) and (20) is taken at position x and time t. The index i is defined modulo 2 and refers to the two possible direction of motion. μ_l and ν_l are random Boolean variables which are responsible for the diffusion and which are also functions of x and t. They are such that $\mu_0=1-\mu_1$ and $\nu_0=1-\nu_1$, so that only one of the two terms of the right-hand side of Eqs. (19) and (20) is selected at a given time and a given position. The Boolean variables μ_l and ν_l take the value 1 with probability p_l. The probability $p_0=1-p_1$ can be adjusted to choose the desired value for the diffusion constant (see Ref. [10] for more details).

R_i is the reaction term. It corresponds to the creation of a particle of type C whenever an A particle and a B particle undergo a head-on collision where they annihilate. k_i is also a Boolean variable which is 1 with probability k. This parameter allows us to tune the reaction rate. For the present simulation we always took k to be equal to 1.

We consider chains of size $2L$, i.e, $x=-L,-L+1,\dots,0,\dots,L-1$. In order to simulate an infinite system, the two extremities of the chain are taken as sources of A and B particles, respectively. The chain should be long enough to ensure that, for the observation time considered, the system is not affected by the boundary conditions.

At time $t=0$, all the sites on the left half of the system are occupied by A particles and, similarly, by B particles on the right half. An equal concentration of A and B ensures that the center of the reaction-diffusion front will stay in the average between $x=-1$ and $x=0$ for all time.

The equations governing the local average density of A and B can be derived in the Boltzmann approximation following the scheme explained in detail in Ref. [10].

We define these averages as

$$\begin{aligned}a(x,t)&=\sum_{i=1,2}\langle a_i(x,t)\rangle \ ,\\ b(x,t)&=\sum_{i=1,2}\langle b_i(x,t)\rangle \ ,\end{aligned} \tag{22}$$

where ⟨ ⟩ represents an average over all the possible realizations of the system. In the spirit of a Boltzmann (or mean-field) approximation, we assume that

$$\langle R_i \rangle \approx k \langle a_i \rangle \langle b_{i+1} \rangle \approx k \frac{ab}{4} \ . \tag{23}$$

This leads [9,10] to the following equation:

$$\partial_t a(x,t) = \frac{\lambda^2}{\tau} \left[\frac{p_0}{2(1-p_0)} \right] \partial_x^2 a(x,t) - \frac{k}{2\tau} a(x,t) b(x,t) \ , \tag{24}$$

and similarly for $b(x,t)$.

Up to irrelevant corrections (see discussion in Ref. [9]), Eq. (24) follows exactly from the microdynamics equations and the Boltzmann assumption. One thus reproduces the equations used by Gálfi and Rácz in which the magnitude of the diffusion constant D and reaction coefficient k have now a microscopic content.

The cellular-automata simulations, using the multispin coding technique [11], were performed on chains of length $L=1024$ and data were collected at ten times between $t=1000$ and 300 000. The values of p_l were chosen as $p_0=\frac{1}{2}$, which corresponds to a diffusion constant $D=\lambda^2/2\tau$.

In order to measure the profile of the region where C is produced, we monitored the number of C particles created at each time step, as a function of the position. Each measurement was performed over a period of $\Delta T = \delta T/10$ steps, where δT is the time interval between two consecutive measurements. This introduces a systematic error of the order of 1% on each measurement, but is necessary to produce sufficient statistics. The data is further averaged over 1792 independent runs.

The width $W(t)$ of the front is defined as

$$W^2(t) = \frac{\sum_{x=-L}^{L-1} (x - \langle x \rangle)^2 R(x,t)}{\sum_{x=-L}^{L-1} R(x,t)} \sim t^{2\alpha} \ , \tag{25}$$

where $\langle x \rangle = x_f$ is the position of the front defined as the first moment of $R(x,t)$.

The time dependence of the width is plotted in Fig. 1. One finds, over $2\frac{1}{2}$ decades (from $t=1000$ to 300 000) the power-law relation

$$W \simeq t^{0.293 \pm 0.005} \ . \tag{26}$$

Also plotted is the width $W_{\langle a \rangle \langle b \rangle}(t)$ of the product $\langle a \rangle \langle b \rangle$ of the averaged densities. Although this differs from W by a factor of order 1, this is found to scale similarly, with relation

$$W_{\langle a \rangle \langle b \rangle} \simeq t^{0.297 \pm 0.005} \ . \tag{27}$$

The error bars are estimated from the analysis of the local gradients. It is plausible that these two exponents are the same, consistently with the predictions of the scaling analysis of the preceding section. This value is not compatible with the prediction of Gálfi and Rácz, namely, $\alpha = \frac{1}{6}$.

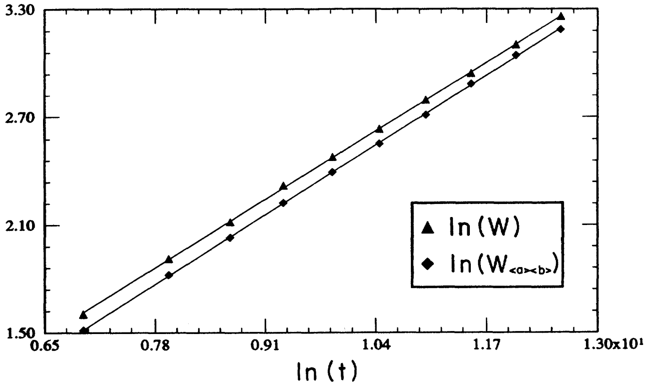

FIG. 1. Logarithm of the width W of the reaction front and logarithm of the width of $R_{\langle a \rangle \langle b \rangle}$ as defined in text, as a function of the logarithm of time.

In Fig. 2, we plot the total reaction rate $N(t)$ and $N_{\langle a \rangle \langle b \rangle}(t)$, which are defined as

$$N(t) = \sum_{x,i} \langle a_i(x,t) b_{i+1}(x,t) \rangle \ ,$$
$$N_{\langle a \rangle \langle b \rangle}(t) = \tfrac{1}{2} \sum_x \langle a(x,t) \rangle \langle b(x,t) \rangle \ , \tag{28}$$

as a function of time on a log-log scale. The straight lines are the best fits.

$$N(t) \simeq t^{-0.498 \pm 0.002} \ , \tag{29}$$

$$N_{\langle a \rangle \langle b \rangle}(t) \simeq t^{-0.134 \pm 0.002} \ . \tag{30}$$

This leads to the following values for the scaling exponents:

$$\alpha = 0.293 \pm 0.005 \ , \tag{31}$$

$$\beta = 0.791 \pm 0.006 \ , \tag{32}$$

$$\gamma = 0.216 \pm 0.003 \ . \tag{33}$$

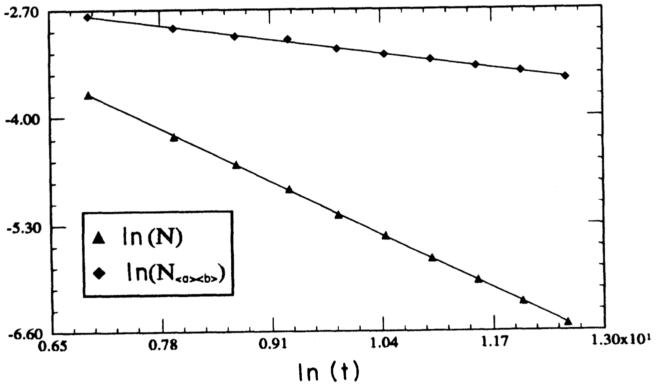

FIG. 2. Logarithm of the total production rate N of C particles and logarithm of $N_{\langle a \rangle \langle b \rangle}$ as defined in text, as a function of the logarithm of time.

This gives us

$$\alpha-\beta+\tfrac{1}{2}=0.002\pm0.002\ , \qquad (34)$$

$$\gamma+2\alpha-\beta=0.011\pm0.006\ , \qquad (35)$$

which compare well with the predictions (9) and (12) of the scaling analysis.

Figures 3 and 4 show the scaling forms for R and R_{MF} with the above values of the exponents, for three typical times. The quality of the scaling fit is very convincing.

IV. MICROSCOPIC ARGUMENTS

The initial problem is very difficult to solve analytically by virtue of the presence of sources of particles and correlations between the species. This makes exact arguments about the behavior of the reaction front very difficult, in contrast to the case of homogeneous reaction-diffusion [1,12]. Nevertheless, we can obtain lower bounds for the range of fluctuations of the reaction by considering a simpler case.

Let us consider a species A, diffusing on a d-dimensional half strip, with a sink formed by a $(d-1)$-dimensional hyperplane perpendicular to the x axis and a source at $x=\infty$. In the long-time limit, the density profile of A, $a(x,t)$, varies like

$$a(x,t)\propto\frac{x}{\sqrt{t}}\ . \qquad (36)$$

Let us introduce a B particle at the origin. This particle performs a random walk, until it meets an A particle, whereupon they annihilate. After moving a distance l the probability of meeting an A particle is approximately

$$P(l,t)\simeq l^d a(l,t)\ . \qquad (37)$$

Therefore the characteristic distance at which a reaction occurs l_r is given by $P(l_r,t)\approx 1$. Hence

$$l_r\simeq t^{1/2(d+1)}\ . \qquad (38)$$

In the true situation, the role of the sink is played by the diffusing B particles. Therefore the sink is effectively diffuse, and so the profile of A particles varies more rapidly. This means that the true reaction width $W(t)$ satisfies

$$W(t)\geq l_r\ . \qquad (39)$$

Another lower bound for the exponent α has been obtained from the scaling analysis (18).

We note first that $d=1$, $l_r\simeq t^{1/4}$, whereas in the mean-field approximation the exponent is $\frac{1}{6}$. This shows clearly that there is a large departure from mean-field behavior. However, a fit of the experimental data to a form $W(t)\simeq t^{1/4}$ is not very convincing. Second, for $d=2$, the fluctuation arguments predict the mean-field exponent. This implies that the upper critical dimension is $d_{\mathrm{up}}=2$. Motivated by analogy with critical phenomena, we have attempted to fit our data for the two-dimensional case [9] with the mean-field exponent plus logarithmic corrections. Figure 5 shows a good fit with a form $W(t)\simeq at^{1/6}\ln(t/t_0)^y+b$, where $y=\frac{1}{2}$. We remark, however, that similar fits with different values of y are equally convincing. The value $y=\frac{1}{2}$ is appropriate to arguments about the largest probable fluctuation in l_r [12,13]. The data for one dimension may in fact be consistent with a form $W(t)\simeq t^{1/4}\ln(t)^{1/2}$ which would also be predicted by such arguments. We emphasize, however, that the fluctuation arguments should only be regarded as giving lower bounds on $W(t)$, and that we therefore have no *a priori* justification for preferring such a fit in one dimension.

For dimensions higher than 2, the fluctuations are

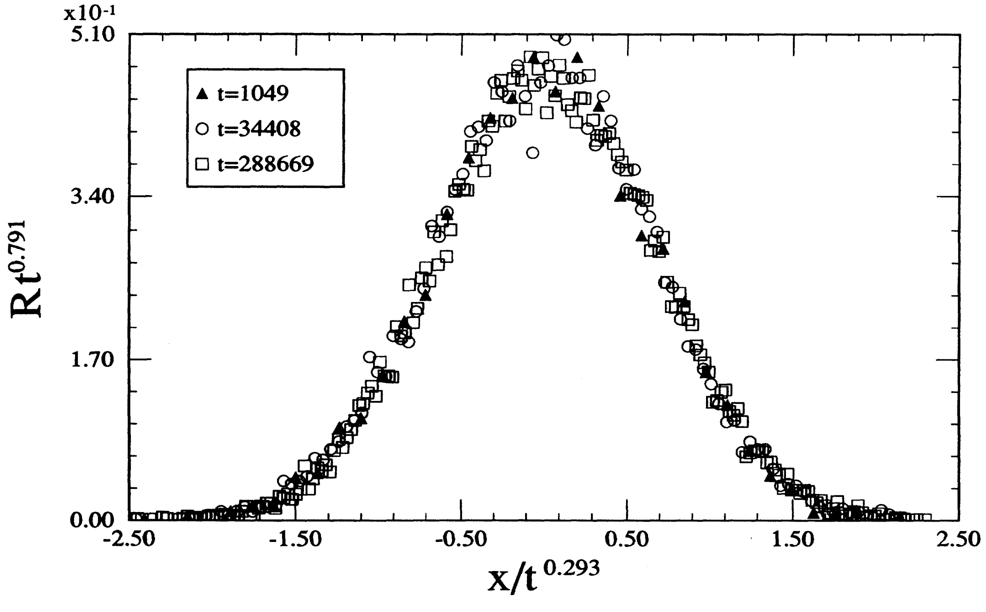

FIG. 3. Scaling profile of the reaction front at three typical times.

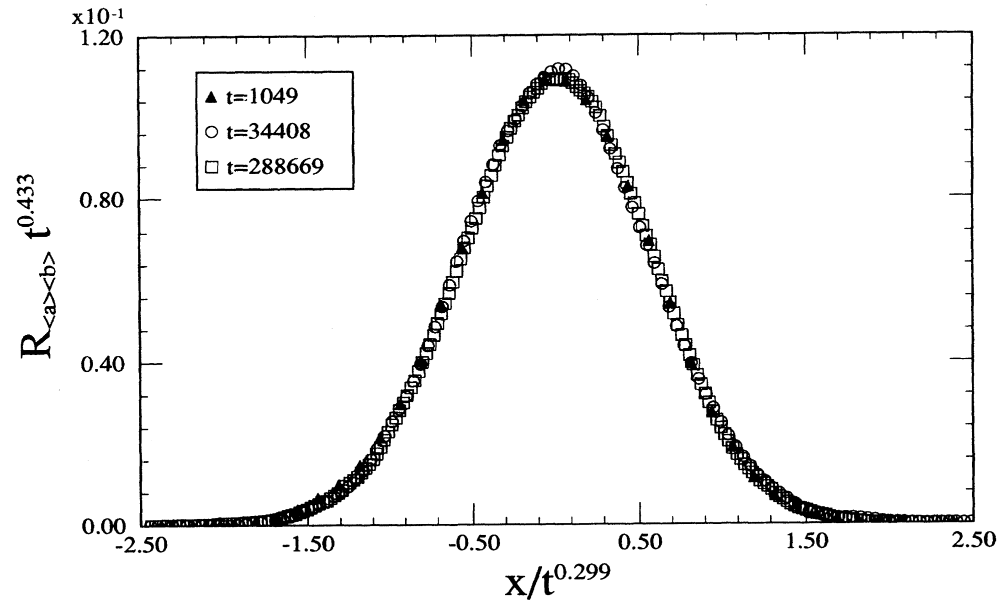

FIG. 4. Scaling profile of $R_{\langle a\rangle\langle b\rangle}$ at three typical times.

shorter in range than the mean-field reaction width. Therefore the depletion of particles drives the reaction front, and so we expect the mean-field predictions to hold.

Similar arguments can be used for the more general reaction $nA+mB\rightarrow C$. For such a reaction to occur, one A particle must meet $(n-1)$ other A atoms and m B atoms within the same volume v, where v is set by the reaction cross section. The probability of such a fluctuation at a distance of order l_r from the center of the front is

$$P_{n,m}=a(l_r,t)^{n-1}b(l_r,t)^m l^d \ . \tag{40}$$

Thus l_r is determined by setting $P_{n,m}\simeq 1$, which gives

$$l_r \simeq t^{(n+m-1)/2(d+n+m-1)} \ . \tag{41}$$

We again recover the mean-field exponent at dimension $d_{\mathrm{up}}=2$. We therefore predict that, assuming that the scaling theory still holds when fluctuations are added, the mean-field exponents hold for dimensions greater than 2, irrespective of the values of n and m.

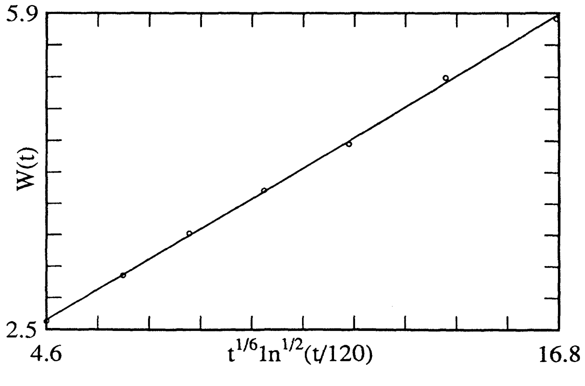

FIG. 5. Fit of the date in Ref. [9] to a mean-field exponent with logarithmic corrections.

V. CONCLUSION

While the arguments presented in the preceding section are not rigorous, they account for the numerical results and give a coherent picture of the phenomenon. In particular, they suggest that the upper critical dimension is $d_{\mathrm{up}}=2$ in this geometry, instead of $d_{\mathrm{up}}=4$ for the homogeneous case.

Another question which is raised by this study is how to add noise in the mean-field reaction-diffusion equations in order to restore the fluctuations. We have considered numerically several attempts in both one and two dimensions, without much success. For instance, the introduction of an additive white-noise term to Eq. (2) led to problems with the positivity of the solutions of the equations. A multiplicative noise, stemming from the idea that the number of particles N in a given volume fluctuates as $\sqrt{N}$, brought the same troubles. Another idea was to take the reaction constant k_1 and k_2 as random variables. This gave well-behaved solutions, but with the mean-field exponents. A value of the reaction constant changing in an *ad hoc* way with time has successfully accounted for the effect of fluctuations in the homogeneous case [14]. There is, however, no *a priori* justification for this approach.

These difficulties call for a first-principles approach. For example, one could consider the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy associated with the CA model, and retain correlation functions of higher order than the one-point functions considered in the mean-field approximation. This method has been used for CA models describing fluids out of equilibrium [15].

Another approach would be to map this classical problem onto a quantum field theory, within which framework one could study the effects of fluctuations using a renormalization-group analysis [16]. Such an approach is under investigation.

The surprising result that the upper critical dimension for reaction fronts appears to be 2, irrespective of the reaction mechanism, needs to be verified numerically. We hope that the discovery of a spectrum of universality classes shall motivate further simulation studies of reaction-diffusion fronts in systems with more complex mechanisms.

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