PHYSICAL REVIEW E **73**, 051109 (2006)

# Reaction-diffusion front width anomalies in disordered media

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(Received 24 August 2005; revised manuscript received 3 February 2006; published 24 May 2006)

We study the front characteristics of the $A+B\rightarrow C$ reaction-diffusion system with initially separated reactants in disordered media, exemplified by two-dimensional (2D) percolation. We investigate the front characteristics as a function of the disorder degree in this system, in particular close to criticality. We show that the front width exponent is *larger* than the mean-field (MF) exponent of 1/6, and at criticality it approaches 1/4, which is the one-dimensional (1D) exponent. We show that previous predictions in the literature for the 2D percolation cluster at criticality are wrong. The results are discussed in the context of other systems with attenuated transport where the front width exponent is *smaller* than the MF exponent. We also study the *short-time* behavior of the front width exponent, and discuss the validity of the scaling relations between the relevant exponents.

DOI: 10.1103/PhysRevE.73.051109 PACS number(s): 82.20.−w, 82.40.−g, 64.60.Ak, 61.43.Hv

## I. INTRODUCTION

Is anomalous diffusion on a regular lattice equivalent to regular diffusion on spatially disordered substrate? It is commonly believed that the answer to this question is positive. In this work we show a case where this is not necessarily true. The system under study is the $A+B\rightarrow C$ reaction-diffusion system with initially separated components on the two-dimensional (2D) percolation system. Following the pioneering work of Gálfi and Rácz (GR) [1], such systems have attracted great interest, both theoretically [2–18] and experimentally [19–22]. When the two reactants are initially separated in space, the reaction takes place in a localized region where the two species meet, exhibiting a dynamic reaction *front* with rich spatio-temporal behavior. At the mean-field (MF) level, this process is described by the reaction-diffusion equations for the $A$ and $B$ concentrations, $\rho_A(x,t)$ and $\rho_B(x,t)$,

$$\dot{\rho}_A = D_A\nabla^2\rho_A - k\rho_A\rho_B,$$

$$\dot{\rho}_B = D_B\nabla^2\rho_B - k\rho_A\rho_B, \qquad (1)$$

where $D_A, D_B$ are the diffusion constants and $k$ is the reaction rate constant. These equations are subject to the initial separation condition

$$\rho_A(x,0) = A_0H(-x), \quad \rho_B(x,0) = B_0H(x), \qquad (2)$$

where $H(x)$ is the Heaviside step function and $A_0$, $B_0$ are the initial densities. This initial condition is one-dimensional, since the separation is along the $x$ coordinate, but the system may be of a higher dimension. The local production rate of $C$ is given by $R(x,t)=k\rho_A\rho_B$, which, asymptotically, is assumed to obey the scaling form [1],

$$R(x,t) = h(t)S\left(\frac{x-x_f(t)}{w(t)}\right),$$

$$w(t) \sim t^{\alpha}, \quad h(t) \sim t^{-\beta}. \qquad (3)$$

In these expressions, distances $x$ are measured relative to the reaction front center $x_f(t)\sim t^{1/2}$, which is the point where $R(x,t)$ is maximal. The width of the front, $w(t)$, is the second moment of $R(x,t)$ and the height $h(t)$ is the value of $R(x,t)$ at $x_f$ (Fig. 1). GR [1] showed that the MF exponents, i.e., above the upper critical dimension $d_c=2$ (Ref. [4]), are

$$\alpha = 1/6, \quad \beta = 2/3. \qquad (4)$$

These results have been confirmed experimentally in a horizontal glass reactor [19] and a gel-free quasi two-dimensional (2D) system [22].

The width exponent $\alpha=1/6$, which was argued [1] to be much smaller than 1/2, the exponent associated with length scales in standard diffusion problems, introduces a new length scale into this problem. Therefore it has been considered in many publications as the basic quantity for characterizing this reaction-diffusion system with initially separated reactants. Below the critical dimension, i.e., for $d<2$, in particular in one dimension (1D), the properties of the reaction front are dominated by fluctuations tending to increase its width. The exact value of the width exponent in 1D has been

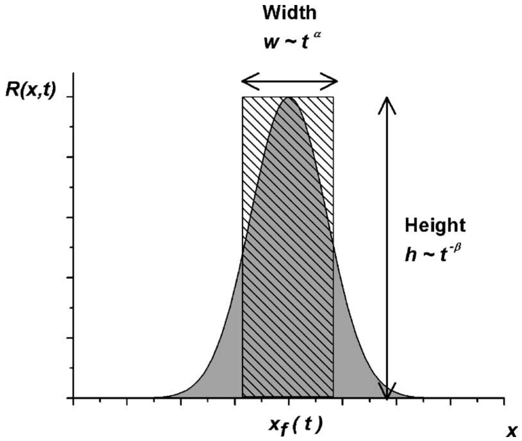

FIG. 1. A schematic sketch of the local production rate $R(x,t)$. The global production rate $R(t)$, which is the integral of $R(x,t)$, is the area under the curve. In regular systems, it can be approximated by the width-height product (shaded rectangle).

the issue of a controversy in the literature [7–12]. However, it is now accepted that the width ($\alpha$) and height ($\beta$) exponents in 1D are

$$\alpha = 1/4, \quad \beta = 3/4. \tag{5}$$

The exponents in Eqs. (4) and (5) obey the scaling relation assumed by GR [1],

$$\alpha - \beta = -1/2, \tag{6}$$

where the right-hand side is the exponent of the global production rate of $C$ in the system, formally defined as $R(t) = \int_{-\infty}^{\infty} R(x,t)dx$. The scaling relation in Eq. (6) results from approximating the production rate within the reaction zone by the multiplication of the width and the height of the reaction front $w(t)h(t)$ (Fig. 1). It is valid when $R(x,t)$ is symmetric around its center, and may not be always valid. Indeed, the above results have been obtained for ordinary diffusion in regular space. However, diffusion in a real system may be avoided in certain points, due to spatial defects or contaminations. It is therefore interesting to find out how such impurities influence the properties of the reaction-diffusion front. Moreover, the theoretical difference between 1D and 2D (MF) may be relevant when a spatial confinement, such as disorder, is introduced into a 2D system. In that case, the dimension may be effectively lower than the critical dimension $d_c = 2$.

Reaction-diffusion fronts in such systems have been partially considered in the literature [3,13–15]. In Refs. [3,14], some results were presented for a fractal substrate, specifically for the infinite percolation cluster in 2D close to criticality. The results were expressed in terms of $d_w$, the anomalous diffusion exponent, defined by the mean displacement of a particle $r \sim t^{1/d_w}$, rather than $r \sim t^{1/2}$ for normal diffusion. For the global production rate $R(t)$, it was argued [3,14] and supported numerically that $R(t) \sim t^{-(1-1/d_w)}$. This follows directly from the time derivative of the total number of products up to time $t$, which is assumed to scale as $t^{1/d_w}$ [14]. In Ref. [3], a prediction was made for the width and the height exponents,

$$\alpha = 1/d_w, \quad \beta = 1. \tag{7}$$

This prediction was based on the numerical values $\alpha = 0.375$ and $\beta = 0.99$, assuming $d_w \approx 2.88$ for this fractal substrate [15]. Combining this result with the result for $R(t)$, one obtains the generalization of Eq. (6),

$$\alpha - \beta = -(1 - 1/d_w). \tag{8}$$

However, the prediction of Eq. (7) has a few drawbacks. First, while the exponent for $R(t)$ [right-hand side of Eq. (8)] gives the correct result in the limit of a regular system, $d_w = 2$, the predictions for $\alpha$ and $\beta$ in Eq. (7) are *inconsistent* with the MF results [Eq. (4)] in this limit. Second, the results were based on only 200 realizations, and, as was pointed out in Ref. [13], more extensive simulations are needed. It should be noted, however, that Eq. (7) does predict qualitatively that the width exponent $\alpha$ for this disordered system is *larger* than the MF value of 1/6.

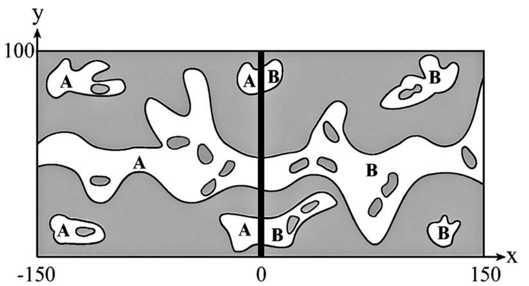

FIG. 2. A schematic sketch of the 2D percolation cluster at criticality with initially separated $A$, $B$ reactants at $t = 0$. Gray regions represent blocked sites. At criticality, one can observe clusters of all sizes, only some of them cross the boundary at $x = 0$.

In this paper we study the 2D percolation system, this time with better averaging and longer times, in the entire range between order and disorder criticality. We also consider a smaller reaction constant, which allows the investigation of the short-time behavior [3] as well. Our results show that the prediction cited in Eq. (7) is *wrong*, and the width exponent is limited by the 1D value of 1/4. This means that the width exponent $\alpha$ is indeed *larger* than the MF value of 1/6, but is limited by 1/4. We discuss this result in the context of attenuated transport in other systems embedded in 2D Euclidean space [23–27]. We also obtain an interesting short-time behavior of $\alpha$.

In the next sections, we first describe in detail our numerical methodology, then present the results for the long- and short-time limits, discuss the validity of the scaling relations, and conclude with several remarks regarding suggested experiments and possible future work.

## II. METHODOLOGY

We carried out a series of Monte Carlo simulations, in a 2D lattice of 300 × 100 sites (Fig. 2). We denote by $p$ the fraction of free sites in the system, so that $q = 1 - p$ is the probability that a given site is blocked. It is well known [26] that when the fraction of free sites is above the critical concentration $p_c = 0.593$ (i.e., $q < 0.407$), percolation through the system is possible. When the free sites concentration is below this value, namely the blocked sites concentration is above the percolation threshold ($q > 0.407$), all the clusters are finite, and percolation between the edges of system is impossible. At criticality, $q = q_c = 0.407$, there is a single infinite cluster, in addition to clusters in all sizes.

The $A$ species were initially placed at $x < 150$ and the $B$ species at $x > 150$. As a result, $A + B$ reaction is possible only in those finite and infinite clusters that cross the boundary at $x = 0$ (Fig. 2). Other isolated clusters of either $A$ or $B$ do not contribute to the reaction process. All possible sites were initially occupied and the reaction constant is just the probability that a reaction occurs when a pair of $A$ and $B$ particles meet. $k = 1$ means a perfect reaction and we chose $k = 0.05$ to allow us to study the short-time behavior, still obtaining the asymptotic behavior in a reasonable time scale ($t > k^{-1}$). The diffusivities of the two species were set arbitrarily to $D_A = 1$, $D_B = 0.5$.

TABLE I. Reaction front exponents, as obtained by simulations with $q=0$, compared to the theoretical MF results for ordered systems, in the short- [3] and long-time [1] limits.

| | $\alpha$ | | $\beta$ | | $\gamma$ | |
|---|---|---|---|---|---|---|
| | Theory | Simulations | Theory | Simulations | Theory | Simulations |
| Short time | 1/2 | 0.519 | Const. | Const. | 1/2 | 0.436 |
| Long time | 1/6 | 0.157 | 2/3 | 0.639 | −1/2 | −0.532 |

In the simulation, each step is split into a diffusion step and a reaction step. In the diffusion step, all the particles in the system are updated sequentially. Each particle has an equal probability to stay in its position or to move to one of the four neighboring sites, regardless whether the site is free or blocked ("blind ant" scheme [24]). If the chosen site is blocked, the particle remains in its previous position. There is no limitation for the number of particles that can occupy a given site. After the entire lattice is updated, the reaction step takes place. If an $A$ particle and a $B$ particle are located in the same site, they react with a probability $k$. When reaction occurs, the $A$ and $B$ particles are removed from the system, and a $C$ particle is created in the same site, contributing to the spatiotemporal production rate $R(x,y;t)$. The $C$ particle is not considered further in the diffusion process, and is assumed not to disturb the diffusion of the $A$ and $B$ particles. The production rate along the $x$ axis, $R(x,t)$, is defined as the sum over all $y$ values,

$$R(x,t) \equiv \sum_{y=-\infty}^{\infty} R(x,y;t). \tag{9}$$

This quantity is the basis for calculating the characteristics of the reaction-diffusion front. The reaction width is the discrete version of the second moment of $R(x,t)$ around the instantaneous position of the front center $x_f$ [2,4], and the height $h(t)$ is simply the value of $R(x,t)$ at $x_f$. In addition, we studied the global production rate $R(t)\sim t^{\gamma}$, which is the integration over $x$ of $R(x,t)$. The exponent $\gamma$ is in fact the generalization of the right-hand side of Eqs. (6) and (8).

The reaction rate $R(x,t)$ was traced at specified times and averaged over 2000 realizations. This number of realizations was determined by analyzing continuously the deviations in the calculated reaction characteristics (width, height, etc.) as the number of realizations is increased. We found that when the number of realizations exceeds 1700, the relative deviation is less than 1%. Therefore, for maximal reliability, the results were averaged over 2000 realizations.

Regarding possible finite-size effects, we claim that the size of our system (300 × 100) can be considered as "infinite" in the studied time region. Regarding the $x$ direction, the active reaction zone (as well as its width) are about 30 times narrower than the system size. Moreover, we found that the initial $A,B$ concentrations near the system edges remain unchanged during the entire simulation time, which is another indication that the system is large enough. Regarding the $y$ direction, we have repeated our calculations for systems of size $300\times300$, as well as $300\times50$. The results are practically the same.

## III. RESULTS

In the limit $q\rightarrow0$, i.e., a regular 2D system, we expect to recover the MF results, since $d_c=2$ is the upper critical dimension. The results for the exponents $\alpha$, $\beta$, $\gamma$ compared with the corresponding theoretical predictions, are summarized in Table I. Details can be found in Ref. [28]. The crossover from the short-time to the asymptotic-time behavior occurs at $t^*\sim k^{-1}\simeq100$ time steps ($k=0.05$). The slight deviations from MF exponents could be attributed to corrections to scaling at the critical dimension. We next studied the exponents $\alpha$, $\beta$, $\gamma$ for increasing values of the blocked sites concentration $q$, *below*, *at*, and *above* $q_c$, in both the short- and long-time regimes.

### A. Long-time limit

The results for the long-time limit are shown in Fig. 3. Each point in these plots is the slope of the log-log plot of the corresponding measure (width, height, etc.) as a function of time for a given $q$. The error of the fit of the log-log plot to a straight line is indicated by the error bars. One can see that the error increases when the disorder increases, due to larger fluctuations in the reaction profiles. Above criticality, i.e., $q>q_c=0.407$, fewer $A$, $B$ clusters around the separation line $x=0$ exist. Hence the diffusion will be significantly suppressed and the reaction front properties will be subject to strong fluctuations. Moreover, above criticality, there exists no infinite cluster, and the reaction occurs only in the finite clusters across $x=0$, until they vanish.

The results shown in Fig. 3 indicate that the exponents remain unchanged for $q\ll q_c=0.407$, i.e., far from criticality, since the reaction is still not affected by the constrained diffusion. Near the critical concentration, usually above $q\simeq0.3$, the exponents $\beta$ and $\gamma$ sharply decrease as the concentration of blocked sites increases, since there is a significant decrease in the feasibility of reaction events in the remaining unblocked sites. The behavior of the width exponent $\alpha$ is more interesting. Above $q\simeq0.3$ it *increases* up to criticality, at criticality $\alpha=0.22$, and above criticality it sharply decreases. The approach to criticality of $\alpha$ can be fitted to a function typical to critical phenomena, $\alpha=\alpha_c+C(q_c-q)^{\nu}$ with $C=-0.186\pm0.087$ and $\nu=0.468\pm0.31$. The value of $\alpha_c$ as obtained from this fit is $\alpha_c=0.229\pm0.021$ which agrees very well with the value obtained at $q=q_c$. Similar functions can be fit to the other exponents [28].

The value of $\alpha\simeq0.22$, obtained close to criticality, is larger than the MF value of 1/6, and close to the 1D results of 1/4. This can be explained by the strong spatial fluctuations in these systems, which result in a much wider reaction

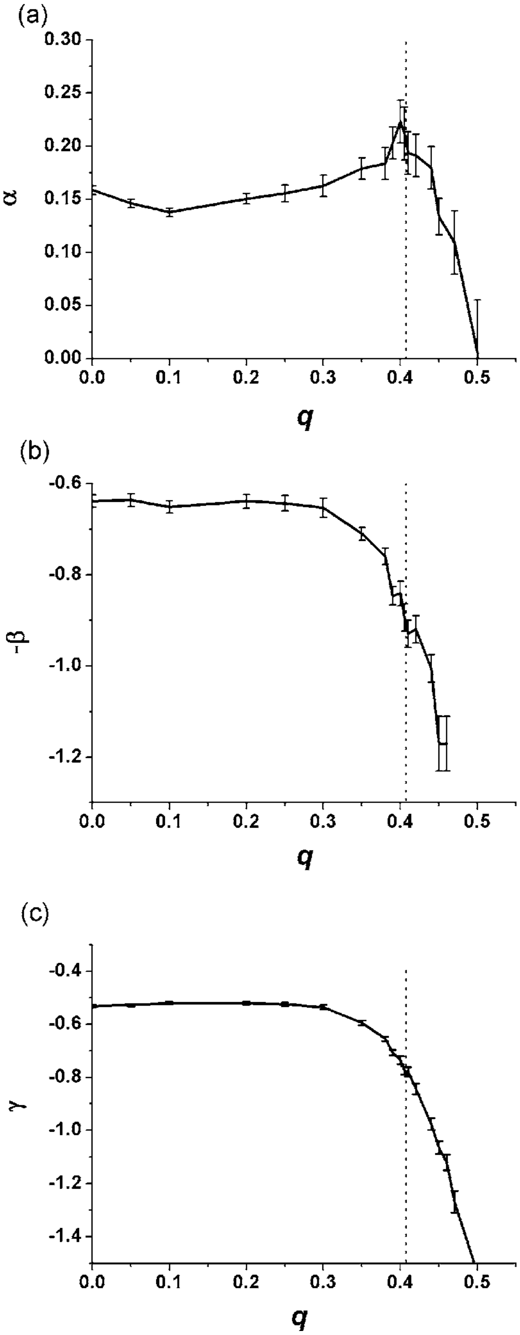

FIG. 3. The reaction front exponents as a function of the fraction of blocked sites $q$ in the long-time regime: (a) Width exponent $\alpha$; (b) Height exponent $\beta$; (c) Global reaction rate exponent $\gamma$. A vertical dashed line is drawn at $q_c=0.407$.

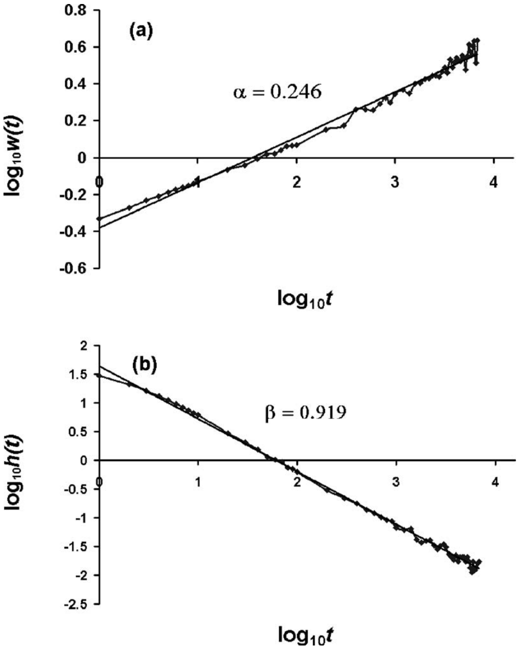

FIG. 4. The width and the height of the reaction front as a function of time, in a log-log scale, for $q=0.405$ (almost at criticality) and $k=1$. (a) The width obeys $w \sim t^{\alpha}$ with $\alpha=0.246$; (b) The height obeys $h \sim t^{-\beta}$ with $\beta=0.919$.

zone than in MF systems, as the reactants must explore distant regions in order to find their counterparts for reaction. This is essentially different from anomalous diffusion (such as subdiffusion) in free space, where the active reaction zone is much more localized. Qualitatively, this conclusion could be drawn from the result $\alpha=0.375$ that was obtained for $q=0.4$ in Ref. [3]. However, our result for $\alpha$, which has been obtained with much better statistics and longer times, *disagrees* with that value. Rather, it indicates that $\alpha$ goes to the 1D result, 1/4, at criticality. This can be explained by observing that at criticality, the average number of free nearest neighbors is about two, such as in 1D, and not four as in free 2D systems. This is another difference between regular diffusion in disordered media and anomalous diffusion in free space.

In order to verify the value of the exponent $\alpha$ at criticality, we have performed another set of simulations, in a larger system ($300 \times 300$), with $q=0.405$ and with a reaction probability $k=1$, where the asymptotic region should set in from the very beginning. The graph of the width $w$ as a function of time (in a log-log scale) is presented in Fig. 4(a). The slope is $\alpha=0.246$ which is very close to the 1D result of 1/4. The simulation was run up to $O(10^4)$ time steps, when the reaction rate has reduced to less than $10^{-3}$ of its maximal value and strong fluctuations show up [as shown in Fig. 4(a)]. However, since the reaction width is a much smaller length scale than the normal diffusion scale, this time limit is considered asymptotic.

The result $\alpha > \alpha_{MF}$ should be discussed in the context of other nonregular systems in 2D. Intuitively, one could expect that any attenuation of the transport will results in a growth of the front *slower* than MF. Several authors [16–18] have investigated the properties of the reaction front subject to various manifestations of anomalous diffusion [23–27]. Araujo [16] studied the case of quenched power-law distribution of transition rates and Koza and Taitelbaum [17] considered the Sinai walk. Recently, Yuste *et al.* [18] replaced the diffusion by subdiffusive motion [27], using a fractional reaction-subdiffusion equation, in which both the motion and the reaction terms are affected by the subdiffusive nature of the process. In all of these works it was found that the width exponent $\alpha$ *decreases* as the anomaly grows, compared

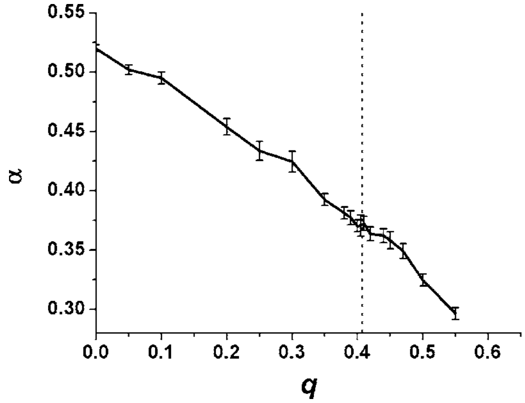

FIG. 5. The reaction front width exponent $\alpha$ as a function of the blocked site fraction $q$, in the *short-time* limit. The exponent decreases monotonically for any $q>0$, reaching the value 0.37 at criticality. A vertical dashed line is drawn at $q_c=0.407$.

to the corresponding normal diffusion. For example, in both works of Araujo [16] and Yuste *et al.* [18], $\alpha=1/3d_w$ ($d\geq 2$), limited by $d_w=2$ for ordinary diffusion. Following these results, one could expect that attenuation of the transport in a system embedded in 2D Euclidean space will have the effect of *decreasing* the front width growth exponent. While this is indeed the case for these systems, it is not the case in others, such as in our system, where the width exponent *increases*. That means that in the context of the reaction-diffusion system with initially separated reactants, there is a difference between attenuation due to anomalous transition rates and anomaly due to a disordered substrate. We find this surprising.

In some sense, this observation resembles the issue raised by Sheu *et al.* [29,30]. They considered two types of reactions, with and without sources, and showed rigorously that for one of the cases, $A+B\rightarrow 0$ in the absence of sources, the asymptotic behavior depends not only on the spectral dimension, which combines inherent transport properties and lattice structure (as commonly believed), but also on the fractal dimension which represents the structure only.

The result for the height exponent $\beta$ at criticality is 0.919, as is shown in Fig. 4(b). This is not very different from the prediction of Ref. [3], $\beta=1$ [Eq. (7)]. For the global reaction rate exponent $\gamma$, we obtained the value −0.722. This is not significantly different from the prediction of Havlin *et al.* [14,15] for this exponent when the entire system (and not only the infinite cluster) is considered,

$$-(1-1/d_w)-(d-d_f)/d_w=-0.653-0.036=-0.689. \qquad (10)$$

In this expression, $d=2$ is the Euclidean dimension and $d_f=91/48$ is the fractal dimension. The difference term, 0.036, is due to the reactions in the finite clusters which cross the $x=0$ line (Fig. 2), but since these reactions are expected to vanish relatively early, the main contribution to the front characteristics is due to the infinite cluster. We found that this difference has a slight, insignificant effect (not more than 0.02), on the results for the width exponent $\alpha$.

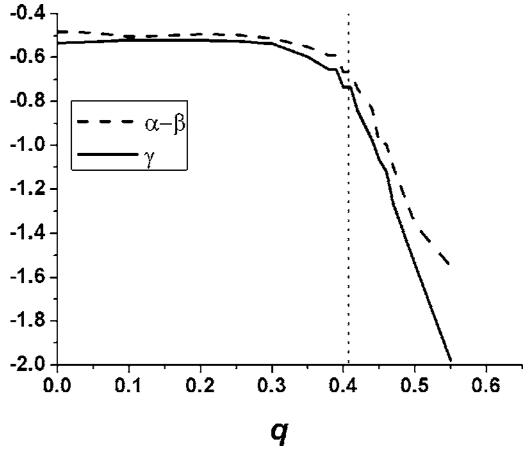

FIG. 6. A comparison of $\alpha-\beta$ and $\gamma$, as a function of $q$. The two values are very close for a small amount of blocked sites, but differ for a concentration of about 0.4 and up. A vertical dashed line is drawn at $q_c=0.407$.

### B. Short-time limit

An interesting behavior of the width exponent $\alpha$ was found in the *short-time* limit, where it is shown (Fig. 5) to decrease monotonically for *any* $q>0$, from its MF value of 1/2 (Ref. [3]) down to the value 0.37 at criticality. In the short-time regime, prior to effective $A$, $B$ mixing, the reaction front properties are governed by *diffusion* [3]. Therefore, the width $w$ is expected to be influenced by any change in the diffusion properties of the system as the blocked sites concentration $q$ increases. As was first shown by Gálfi and Rácz [1], it is only in the long-time regime that the width $w$ is affected also by the reaction, resulting in a new length scale with a new exponent 1/6. Similar findings have been obtained for the behavior of the location of the reaction front center $x_f$ (Ref. [28]), which is the other length scale in the problem.

### C. Scaling relations

Back to the long-time limit, another issue to be looked at is the validity of the relation $\alpha-\beta=\gamma$ [Eqs. (6) and (8)], first suggested by Gálfi and Rácz [1]. For $q=0$ this relation is obeyed within an error of 10%, $\alpha-\beta=0.157-0.639=-0.482$, whereas $\gamma=-0.532$. We next look at the validity of this relation as a function of the blocked sites concentration $q$, by comparing the value of $\alpha-\beta$ with $\gamma$. The results are presented in Fig. 6. Far from criticality, the value of $\alpha-\beta$ is very close to the value of $\gamma$. Close to the critical value of $q_c=0.407$, the difference between the two values increases, and above $q_c$ the relation seems not to be valid anymore. For example, for $q=0.55$ the deviation from the predicted relation reaches 21%.

The physical interpretation of this deviation is as follows. As mentioned above, the $\alpha-\beta=\gamma$ relation is derived from the approximate assumption that the multiplication of the width and the maximal height behaves as the overall area under the function (Fig. 1). This assumption implies that the function behaves "normally," i.e., no infinite tails and no sharp decrease around the maximum. In a disordered system, this assumption is not necessarily valid. Due to the suppression of diffusion, the reaction around the front is active much longer than the reaction in regions far from the initial boundary $x=0$ (Fig. 2). As a result of this localization, the relation $\alpha-\beta=\gamma$, which was shown to be valid for ordered systems, breaks down as the disorder concentration reaches criticality. Figure 6 shows that $\alpha-\beta>\gamma$ (namely $|\alpha-\beta|<|\gamma|$), which confirms that the global reaction rate in the entire system (represented by $\gamma$) decays faster in time than the reaction rate in the vicinity of the front (represented by $\alpha-\beta$). This implies that in this region one cannot estimate the width and height exponents based on the global reaction rate exponent.

## IV. SUMMARY

In summary, we studied the influence of quenched disorder on the reaction front exponents in the 2D percolation system, for the entire range of disorder concentration $q$, from complete order ($q=0$) to criticality ($q=0.407$) and beyond. We showed the change of the scaling exponents above $q\simeq 0.3$ in the long-time limit, and the change of the width exponent $\alpha$ for *any* $q$ in the short-time limit. Near the percolation threshold, the system exhibits 1D features. In particular, the front width exponent in the long-time limit is $\alpha=0.246$, which is very close to the 1D result of 1/4. This result contradicts earlier results in the literature, and demonstrates that in disordered media, below the critical dimension $d_c=2$, the width exponent is *larger* than in MF.

The experimental system of Park *et al.* [22], which is quasi-2D, can be used to test our predictions. This system consists of two microscope slides with dimensions 75 mm × 25 mm and a gap of 150 $\mu$m in between. The spatial confinement resulting from the narrow gap allows normal diffusion, rather than convection, even without a gel. The exponents of the front characteristics were found to agree with the MF results within the experimental error. In order to study experimentally our findings, one can add impurities to this system in order to create a 2D percolation system. Another possible future work is to consider *dynamic* disorder, which changes in time during the process. For example, the product particles ($C$) may dynamically block the diffusion of the $A$ and $B$ particles so that the disorder increases in time.

## ACKNOWLEDGMENTS

We thank David A. Kessler for useful discussions and Moti Havlin and Ahikam Ben-Avraham for computational assistance. We thank Katja Lindenberg for bringing Refs. [29,30] to our attention. This research was supported by the Israel Science Foundation (Grant No. 1342/04).

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