Reaction-diffusion front in a system with strong quenched disorder

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Using the Sinai model, we study the effect imposed by strong quenched disorder on the dynamic properties of the reaction front formed in $A+B\rightarrow C$ reaction-diffusion systems with initially separated reactants. We confirm that the general scaling ansatz is valid also for disordered systems and find that a single characteristic length controls the asymptotic properties of the entire system. We compare our results with those obtained for different types of disorder as well as with results derived for a translationally invariant space. [S1063-651X(97)09111-3]

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I. INTRODUCTION

It is by now well established that kinetics of diffusion-limited reactions in random and/or confined geometries is qualitatively different from that observed in a homogeneous three-dimensional space [1]. The singular properties of such systems arise most often from the absence of a mechanism that would enable thorough (local) mixing of the reactants. There can be many physical reasons for inefficiency of the mixing, e.g., the absence of convective stirring in or on solid, viscous or porous media [1], concentration fluctuations in the initial condition in homogeneous low-dimensional spaces [2–5], or anomalous transport properties in disordered media [1,6–9], but its most significant consequence is that the local reaction rate is not proportional to the product of the mean local concentrations of the reactants, which renders the commonly used mean-field approximation generally inappropriate for such systems.

There are many examples of chemical and biological diffusion-controlled reactions as well as many ''nonchemical'' processes resembling them, e.g., exciton-exciton, defect-defect, soliton-antisoliton and electron-hole recombinations. In many cases the reactants are initially separated and as time goes on, owing to diffusion, they start to mix and react. However, the theoretical and the experimental research in this field have concentrated mainly on translationally invariant spaces [10–21], a condition that in reality is often violated.

The asymptotic properties of the $A+B\rightarrow C$ reaction front in homogeneous media attracted much interest in the past decade. It was found that if A's and B's initially occupy opposite sides of the $x=0$ plane, the mean local reaction rate $R(x,t)$ asymptotically assumes, at location x and time t, the scaling form [11,12]

$$R(x,t)=h(t)S_R\left(\frac{x-x_f(t)}{w(t)}\right), \tag{1}$$

where $x_f(t)\propto t^{1/2}$ is the point at which R attains its maximal value and the width w and height h of the reaction front take on simple forms

$$w(t)\propto t^{\alpha}, \quad h(t)\propto t^{-\beta}, \tag{2}$$

with $0\leqslant\alpha<\frac{1}{2}$ and $\beta=\alpha+\frac{1}{2}$. Although it was later suggested [13,14] that the simple power-law dependence in Eq. (2) should be modified, for space dimensions $d\leqslant 2$, by logarithmic factors, the exponent α remains the basic quantity describing the reaction front at long times. For $d\geqslant 2$ it was found that $\alpha=1/6$ if both A's and B's are mobile [15,16] and $\alpha=0$ if one of the diffusion constants vanishes [17,18]. In one-dimensional systems the properties of the reaction front turned out to be dominated by fluctuations tending to increase its width, and most researchers agree that because of them α takes on a value 1/4 [13–16,19,20].

The influence of quenched local disorder on the reaction interface was recently studied by Araujo [9], who considered random walks on a d-dimensional lattice ($d=1,2,3$). The quenched disorder was modeled by assigning to each lattice site j a random disorder variable $0<\tau_j<1$ taken from a power-law distribution $p(\tau_j)=\nu\tau_j^{\nu-1}$, with disorder strength $0<\nu<1$ fixed for the whole lattice; the set $\{\tau_j\}$ modifies the jump rates of the random walkers by requiring that the mean time spent by them at site j before leaving it for one of its nearest neighbors is proportional to $1/\tau_j$. That study confirmed the validity of Eqs. (1) and (2) with $\alpha=\nu/(1+3\nu)$ for $d=1$ and $\alpha=\nu/6$ for $d\geqslant 2$ and β related to α through

$$\beta=\alpha+1-1/d_w, \tag{3}$$

where [6]

$$d_w=\begin{cases}(1+\nu)/\nu, & d<2\\ 2/\nu, & d\geqslant 2\end{cases} \tag{4}$$

is the anomalous diffusion exponent related to the scaling properties of the mean-square displacement of noninteracting particles diffusing in this system $\langle x^2\rangle\sim t^{2/d_w}$.

In this paper we investigate the properties of reaction fronts under the so-called Sinai disorder [22], which has been suggested as being relevant to various physical phenomena, e.g., dynamics of dislocations in doped crystals, slow dynamics of random-field magnets [23], $1/f$ noise [24], transport in amorphous or porous media [25], and charge separation in photosynthetic systems [26]. In the Sinai model one considers a one-dimensional lattice in which to each of its sites a local bias field $-1<E_j<1$ is assigned. The set

$\{E_j\}$ consists of independent random variables drawn from a given probability distribution $\rho(E)$. The two jump probabilities at site j, P_j^+, that a random walker will hop from j to $j+1$, and $P_j^- = 1 - P_j^+$ that it will jump to $j-1$, are related to E_j by

$$P_j^+ = \tfrac{1}{2}(1+E_j), \quad P_j^- = \tfrac{1}{2}(1-E_j). \tag{5}$$

The motion of a random walker in such a medium corresponds to diffusion in the presence of a potential field

$$V_j = \sum_{k=j_{\min}}^{j} \ln(P_k^+/P_k^-), \tag{6}$$

where $j_{\min}$ is the first node of a finite lattice (for infinite systems this definition requires suitable rescaling to avoid divergent sums). This potential, being a sum of random variables, can be regarded as performing a random walk itself. Sinai [22] proved rigorously that if $\langle \ln(P_j^+/P_j^-)\rangle = 0$ (which means that there is no global bias in the system) and if $0 < \langle \ln^2(P_j^+/P_j^-)\rangle < \infty$, then the mean-square displacement of a random walker grows extremely slowly

$$\langle x^2(t)\rangle \propto \ln^4 t. \tag{7}$$

This remarkable result is caused by stretches of bias with the same direction that tend to confine the particles inside potential wells.

The aim of our study is to examine the impact of this type of quenched local disorder on the asymptotic, long-time properties of the reaction front formed in the $A+B \rightarrow C$ reaction-diffusion system with initially separated reactants. In addition to the Sinai type of disorder, we will assume that the initial concentrations of A's and B's are the same, their jumping rates depend on the same set of local bias variables $\{E_j\}$, and that upon collision of unlike particles a reaction is certain to occur. We shall focus our attention on averages over disorder realizations: $x_f(t)$, $w(t)$, $h(t)$, and $R(x,t)$. In particular, we confirm the validity of the general scaling ansatz (1) with $w(t) \propto \ln^2 t$, $h(t) \propto (t \ln t)^{-1}$, and the total reaction rate $R(t) \equiv \int R(x,t)dx \propto \ln t/t$. The Sinai disorder turns out to be so strong that the reaction has practically no impact on the interface and the asymptotic properties of the local reaction rate and concentration profiles of A's and B's are governed by a *single* characteristic length scale $\sqrt{\langle x^2(t)\rangle} \propto \ln^2 t$ both inside and outside the reaction zone.

II. ANALYSIS

Since we assume that the mechanical properties of A's and B's are the same and the initial condition is symmetric with respect to $x=0$, the mean position of the reaction front center does not change in time

$$x_f(t) = 0. \tag{8}$$

The mean total reaction rate $R(t)$ is equal to $dI(t)/dt$, where $I(t)$ is the mean cumulative number of reactions that have occurred by time t. This in turn is proportional to the typical distance traveled by particles during time t, i.e., to $\sqrt{\langle x^2\rangle}$. Thus, asymptotically,

$$R(t) = \frac{dI(t)}{dt} \propto \frac{d\sqrt{\langle x^2\rangle}}{dt}. \tag{9}$$

Taking into account Eq. (7), we thus find that

$$I(t) \propto \ln^2 t, \quad R(t) \propto \frac{\ln t}{t}. \tag{10}$$

Assuming the validity of the scaling ansatz (1), $R(t) = \int R(x,t)dx$ should be proportional to the product of the width and height of the reaction front. Using Eq. (10), we thus conclude that

$$w(t)h(t) \propto R(t) \propto \frac{\ln t}{t}. \tag{11}$$

The width of the reaction front is usually defined as the second moment of the local reaction rate [11]

$$w^2(t) = \frac{\int_{-\infty}^{\infty} x^2 R(x,t)dx}{\int_{-\infty}^{\infty} R(x,t)dx}, \tag{12}$$

which is consistent with the scaling ansatz (1) (see Ref. [27] for thorough discussion of this topic). This quantity is bounded from above by the rms displacement of a noninteracting random walker, i.e., by the quantity expected to be the measure of the width of the interfacial region between A's and B's in the absence of reaction. Indeed, comparing two similar systems, one with and the other without a reaction, we can see that the reaction can only decrease the width of the interfacial layer. This stems from the fact that the further a particle diffuses into the region initially occupied by the unlike species, the greater probability that it meets a particle of the ''opposite'' type and reacts. Such a decrease in the width of the interface due to a reaction was observed in homogeneous media, where α was found to be less than $\frac{1}{2}$, in the study of disordered system by Araujo [9], and in a one-dimensional reaction-diffusion system with Lévy flights [10]. We thus arrive at a general inequality

$$w(t) \leqslant \sqrt{\langle x^2\rangle}, \tag{13}$$

which, due to Eq. (7), in the case of Sinai systems asymptotically takes on a form

$$w(t) \leqslant C_0 \ln^2 t, \tag{14}$$

where $C_0 > 0$ is a constant.

Since we assume that upon collision of unlike particles a reaction is certain to occur, in our model particles A and B remain separated *for all times*. The reaction front in such a system is conveniently analyzed with a help of two quantities [21]: the distance between the closest particles A and B, $l_{AB}(t)$, and the location of the midpoint (''center of mass'') between these particles $m(t)$. Whenever there is a reaction in the system, $l_{AB} = 0$ and its location is given by m. These two quantities describe properties of a *particular* realization of the system and should be carefully distinguished from the quantities employed in the scaling ansatz (1), the latter rep-

resenting averages over the entire *ensemble* of different bias fields $\{E_j\}$ and different thermal histories of diffusing particles.

To find the lower bound for $w(t)$ let us for a moment assume that we monitor only those reactions that occur immediately after a previous reaction happened at the origin of the system $x=0$. Let $w^*(t)$ denote the width of the reaction front computed for such reactions and let $l^*_{AB}(t)$ denote the closest A-B distance calculated just after a pair A-B has reacted at $x=0$. If a reaction has just occurred at $x=0$, the next collision of A and B will most likely take place somewhere between the current locations of the rightmost A and leftmost B particles. Therefore, $w^*(t)\propto\langle l^*_{AB}(t)\rangle$. However, if we choose to monitor all reactions, we shall find that hardly ever do they follow a reaction precisely at $x=0$ simply because the reaction front is relatively broad. The location of the midpoint $m(t)$ fluctuates around its mean value 0 and this random wandering can only increase the width of the reaction front as compared to our reference system where a reaction is taken into account only after a pair A-B collides at $x=0$. Therefore we expect that $w(t)\geqslant w^*(t)$. Notice now that $\langle l^*_{AB}(t)\rangle\propto\langle l_{\rm trap}(t)\rangle$, where $l_{\rm trap}$ is a quantity extensively studied in the so-called trapping problem [28,29]: A perfect immobile trap is sitting at $x=0$ and a swarm of diffusing particles, initially uniformly, occupies the whole system. The length $\langle l_{\rm trap}(t)\rangle$ is defined as the average distance between the trap and the closest particle at time t and, following our analysis, satisfies $w(t)\geqslant w^*(t)\propto\langle l^*_{AB}(t)\rangle\propto\langle l_{\rm trap}(t)\rangle$ and therefore constitutes the lower bound for $w(t)$,

$$w(t)\geqslant C_1\langle l_{\rm trap}(t)\rangle, \tag{15}$$

with $C_1>0$ being another constant. The properties of $\langle l_{\rm trap}(t)\rangle$ were already studied for several models, including homogeneous systems [29–31], the Sinai model [32], and the random waiting time disorder [32]. It was rigorously proven [29] that in the absence of disorder $\langle l_{\rm trap}(t)\rangle\propto t^{1/4}$, which is in accordance with Eq. (15). The $\langle l_{\rm trap}(t)\rangle$ distance in the Sinai model was found to read

$$\langle l_{\rm trap}(t)\rangle\propto\ln^2 t. \tag{16}$$

It now follows from Eqs. (11)–(16) that asymptotically

$$w(t)\propto\ln^2 t, \quad h(t)\propto(t\ln t)^{-1}, \tag{17}$$

which, together with Eq. (10), constitutes the main result of this section.

III. NUMERICAL SIMULATIONS

To support our conjecture we have performed extensive numerical simulations. We used a one-dimensional lattice with $L=801$ sites, initially putting particles A (and B) to the left (right) of its center with concentration $a_0=b_0=1.0$. For simplicity we chose as the distribution of the local bias variables E_j,

$$\rho(E_j)=\tfrac{1}{2}[\,\delta(E_j-E)+\delta(E_j+E)\,], \quad E=3/4. \tag{18}$$

This choice corresponds to a binomial distribution $\mathrm{Prob}(P_j^+=7/8)=\mathrm{Prob}(P_j^+=1/8)=1/2$. Particles of the same type do not interact among themselves. When two unlike particles collide, they react and are replaced by a C. Particles C, or the reaction products, are immobile, chemically inert, and do not interact in any way with A's and B's. There is no exclusion principle. Taking into account large fluctuations and slow convergence of the Sinai model to the long-time limit, we averaged the results of our simulations over $N=13\,000$ configurations of the bias field $\{E_j\}$ and for maximal times $t_{\rm max}=10^7$ steps per particle.

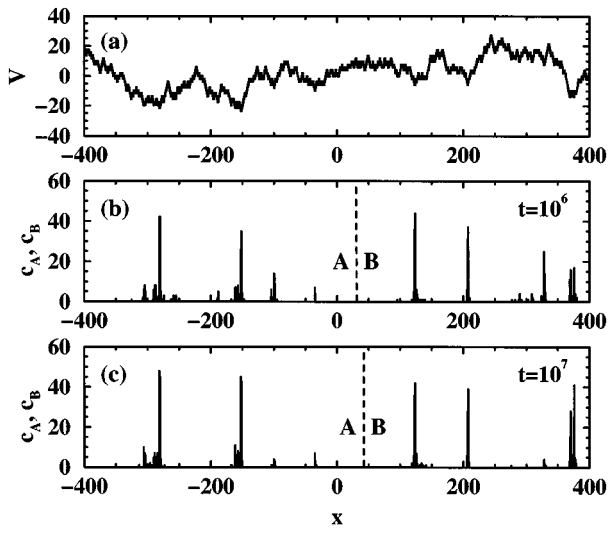

FIG. 1. (a) Sinai potential $V(x)$ for a typical realization of the bias field $\{E(x)\}$; (b) the corresponding concentration profiles $c_A(x,t)$ and $c_B(x,t)$ of A and B, respectively, at $t=10^6$; (c) the same as in (b) but for $t=10^7$. The vertical dashed lines represent the midpoint $m(t)$ separating A from B; $m(10^6)=32$ and $m(10^7)=43$. The distances between the closest A and B are $l_{AB}(10^6)=131$ and $l_{AB}(10^7)=152$.

In Fig. 1(a) we present a plot of the Sinai potential (6) for a typical realization of the local bias field $\{E(x)\}, x=-400,\ldots,400$. The corresponding concentrations of particles A and B at times $t=10^6$ and $t=10^7$ are depicted in Figs. 1(b) and 1(c), respectively. In this particular case particles $A(B)$ were found to the left (right) of $m(t)=32$ for $t=10^6$ and $m(t)=43$ for $t=10^7$, and the distance between the closest A and B was equal to $l_{AB}(10^6)=131$ and $l_{AB}(10^7)=152$, respectively. We can see in these figures the main properties of the Sinai model. Most of the particles are being captured at the bottom of potential wells, with very small probability to get out of them, a phenomenon leading to the extremely slow transport through such a medium. As time goes on, the particles eventually manage to escape from shallow wells and are getting ''trapped'' by a few particularly deep potential minima that dominate the long-time properties of the system [7,25]. Consequently, very few reactions are being recorded during a single simulation (on average we observed only 48 reactions per a single run of 10^7 steps). It is also clear that the behavior of the Sinai system is *very* sensitive to the particular realization of the local field $\{E_j\}$, and the results of simulations are very ''noisy.'' Therefore, a large number of runs is required before reasonably good averages can be obtained.

Because of the extremely slow reaction rate, direct verification of the scaling ansatz (1) is practically impossible. In-

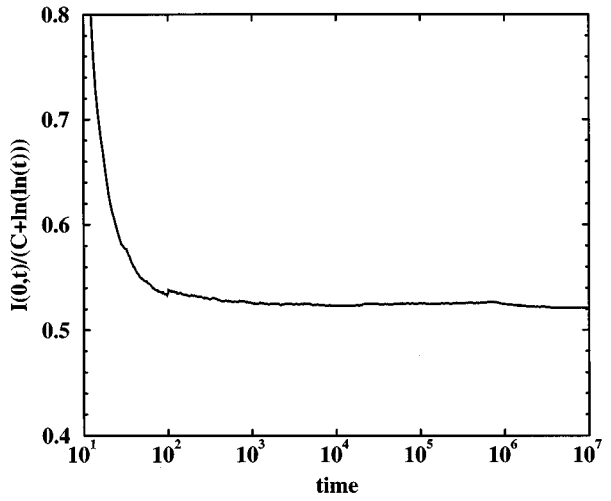

FIG. 2. $I(0,t)/[C_I+\ln(\ln t)]$ as a function of time t, with $C_I=-0.62$.

stead, we have checked whether the cumulative number of reactions that have occurred at x by time t, $I(x,t)=\int_0^t R(x,\tau)d\tau$, takes on the scaling form consistent with Eqs. (1) and (17),

$$I(x,t)\propto[C_I+\ln(\ln t)]G\left(\frac{x}{\ln^2 t}\right). \tag{19}$$

Here C_I may be interpreted as the integration constant, but actually we introduced it to compensate for our arbitrary choice of the base of the logarithm functions used in Eq. (19). To estimate its value we fitted $I(0,t)\propto C_I+\ln(\ln t)$ to our simulation data for the cumulative number of reactions at the origin of the system. Using the data for $t\geqslant 10^2$ we found that the best fit corresponds to $C_I\approx -0.62$, which is the value we shall use henceforth. In Fig. 2 we present the plot of $I(0,t)/[C_I+\ln(\ln t)]$ as a function of time. This function turns out to be constant to a good approximation for $t>10^3$, i.e., in the region where we expect the asymptotic Sinai behavior (7) to be valid.

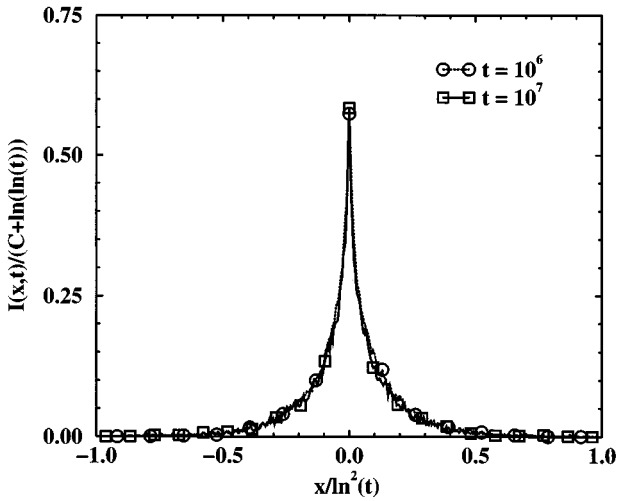

FIG. 3. Scaling plot of $I(x,t)/[C_I+\ln(\ln t)]$ as a function of $x/\ln^2 t$ for $t=10^6$ (dashed line and circles) and $t=10^7$ (solid line and squares), with $C_I=-0.62$.

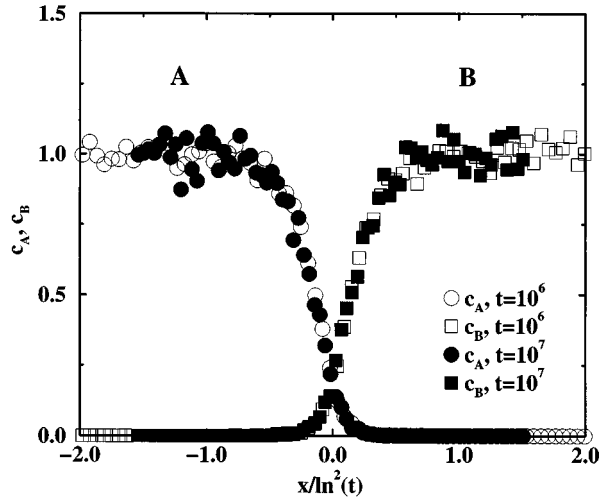

FIG. 4. Concentrations c_A (circles) and c_B (squares) of A and B, respectively, as functions of the scaling variable $x/\ln^2 t$ for $t=10^6$ (empty symbols) and $t=10^7$ (filled symbols).

Figure 3 depicts the scaling plot of $I(x,t)/[C_I+\ln(\ln t)]$ as a function of $x/\ln^2 t$ for $t=10^6$ (dashed line and circles) and $t=10^7$ (solid line and squares). The two curves match each other remarkably well. A corresponding scaling plot for the mean concentrations $c_A(x,t)$ and $c_B(x,t)$ of particles A and B, respectively, is shown in Fig. 4. These two plots confirm the validity of the scaling ansatz for $R(x,t)$, $c_A(x,t)$, and $c_B(x,t)$.

Finally, in Fig. 5 we present a log-log plot of three quantities: $\langle l_{AB}(t)\rangle$, $w(t)$, and the second moment of $m(t)$, $m_2(t)\equiv\sqrt{\langle m^2(t)\rangle}$, as functions of $\log_{10}t$. The mean slopes of these curves, calculated for $t>10^3$, are 2.2, 2.2, and 2.1, respectively. Taking into account a very slow convergence of the Sinai model to its asymptotic time limit and a possible effect of atypical configurations [25], the agreement with our conjecture (17) that $w(t)\propto\ln^2 t$ is remarkably good. Similarly, these data suggest that $\langle l_{AB}(t)\rangle$ and $\sqrt{\langle m^2(t)\rangle}$ are also asymptotically proportional to $\ln^2 t$.

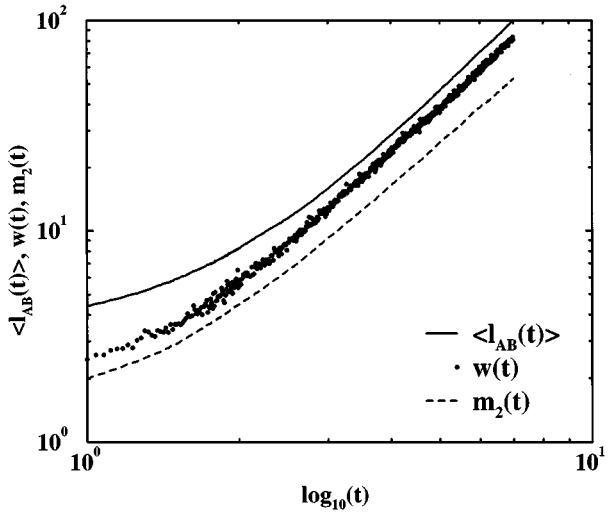

FIG. 5. The log-log plot of $\langle l_{AB}(t)\rangle$ (solid), $w(t)$ (circles), and $m_2(t)\equiv\sqrt{\langle m^2(t)\rangle}$ (dashed) as functions of $\log_{10}t$.

IV. DISCUSSION AND CONCLUSIONS

In purely diffusive systems ($R=0$) the spatiotemporal evolution of concentration profiles c_A and c_B is governed by a single characteristic length, the rms displacement of noninteracting particles $\sqrt{\langle x^2(t)\rangle}$. In reaction-diffusion systems ($R>0$) this property holds only for the regions lying far from the reaction interface; these two areas are dominated by a single species (A or B) so that R is negligibly small there. However, the reaction plays a crucial role inside the reaction zone and its typical effect is to significantly decrease the width of the interfacial layer, so that the dynamics of the interface is governed by a new characteristic length $w(t)$ measuring the width of the reaction front. Consequently, the concentration profiles $c_A(x,t)$ and $c_B(x,t)$ assume *different* scaling forms *outside* and *inside* the reaction zone. These findings were confirmed both in homogeneous [11,27,33] and some random [9] media. A surprising conclusion emerging from our present study is that Sinai disorder is so strong that the reaction has practically no impact on the interface and the entire system is governed by a *single* characteristic length scale $\sqrt{\langle x^2(t)\rangle}\propto \ln^2 t$. Consequently, c_A, c_B, and $R(x,t)$ asymptotically assume the scaling forms

$$c_A(x,t)\propto S_A(x/\ln^2 t)\,,$$

$$c_B(x,t)\propto S_B(x/\ln^2 t)\,,$$

$$R(x,t)\propto (t\ln t)^{-1} S_R(x/\ln^2 t)\,, \tag{20}$$

valid for *any* x. This phenomenon can be clearly appreciated in Figs. 3 and 4, where the scaling encompasses regions lying both inside and outside the reaction zone whose width is approximately equal to $0.3\ln^2 t$. Note also that the above scaling forms imply that the mean local reaction rate $R(x,t)$ cannot be expressed as a function of $c_A(x,t)$ and $c_B(x,t)$ only, a clear indication that the reaction front under Sinai disorder cannot be investigated by mean-field methods.

Slow reactions are known to modify the short-time behavior of reaction-diffusion systems, but in the long-time limit their effect on the reaction front is similar to that of fast reactions [34,35]. Therefore, although this study pertains only to fast (instantaneous) reactions, we expect our main (asymptotic) results to be valid for slow reactions too.

The rms displacement of noninteracting particles $\sqrt{\langle x^2(t)\rangle}$ and the reaction width $w(t)$ are related to each other through inequality (13). In addition to controlling the asymptotic properties of the concentration profiles, these two characteristic length scales govern, via Eqs. (9) and (11), the long-time behavior of two other important quantities: the height of the reaction front $h(t)$ and the total reaction rate $R(t)$. Our analysis shows also the relevance of the third characteristic length $\langle l_{\rm trap}(t)\rangle$, which constitutes the lower bound for $w(t)$, but is easier to investigate.

TABLE I. Asymptotic properties of the rms displacement of a single particle diffusing in a given medium $\sqrt{\langle x^2(t)\rangle}$, the width of the reaction front $w(t)$ in $A+B\to 0$ systems, and the mean nearest-neighbor distance $\langle l_{\rm trap}(t)\rangle$ in the trapping problem for three different types of disorder in one dimension. The data for $w(t)$ and $\sqrt{\langle x^2(t)\rangle}$ come from [13] (homogeneous medium) and [9] (random waiting times). The data for $\langle l_{\rm trap}(t)\rangle$ are from [29,32]. The parameter ν is the disorder strength of Ref. [9].

Length	No disorder	Random waiting times	Sinai disorder
$\sqrt{\langle x^2(t)\rangle}$	$t^{1/2}$	$t^{\nu/(1+\nu)}$	$\ln^2 t$
$w(t)$	$t^{1/4}(\ln t)^{1/2}$	$t^{\nu/(1+3\nu)}$	$\ln^2 t$
$\langle l_{\rm trap}(t)\rangle$	$t^{1/4}$	$t^{\nu/(1+3\nu)}$	$\ln^2 t$

In Table I we compare these three major characteristic lengths, related to essentially different properties of the medium, for various types of disorder in one dimension. The data confirm our conjectures (13) and (15). Note that, except for a small logarithmic correction to $w(t)$ in a translationally invariant space, the asymptotic forms of $w(t)$ and $\langle l_{\rm trap}(t)\rangle$ turn out to be the same. It was recently suggested [32] that $\langle l_{\rm trap}(t)\rangle$ is asymptotically proportional to the rms displacement of particles in systems with hard-core interactions $\sqrt{\langle x^2(t)\rangle_{\rm HC}}$. We thus conclude that both $\langle l_{\rm trap}(t)\rangle$ and $\sqrt{\langle x^2(t)\rangle_{\rm HC}}$ serve as very good approximations of $w(t)$ in one dimension. We also suggest that reaction-diffusion systems with initially separated reactants are characterized by a single length scale only in such (random) media in which the leading asymptotic behavior of the rms displacement with or without hard-core interactions is the same, which is the case in Sinai disorder [32,36].

ACKNOWLEDGMENTS

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