Random walks and reactions on dendrimer structures

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Abstract

We perform model calculations of individual particle random walks on dendrimer structures, due to the emerging interest and wealth of recent experimental data. We use varying coordination numbers and generation orders. We find that it is practically impossible to verify numerically the customary walk properties with the existing asymptotic formalism, because of finite-size effects, even at short times and large system sizes. Several different sets of boundary conditions are employed, producing large differences in the observed behavior for the number of sites visited at least once, as a function of time, S_N. We also investigate the case of reacting (annihilating) particles, using the well-known $A + A$ and $A + B$ models, in which case we find an anomalous scaling for the decay of the particle density, with the scaling exponents being $f = 0.80 \pm 0.03$ and 0.50 ± 0.03, respectively. We discuss how diffusion in dendrimers is analogous to biased random walks in one dimension, which demonstrates a breakdown of the usual scaling between the reaction progress (or reactant survival) and S_N. An appendix gives results for biased walks in one dimension. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

The advent of a new class of structurally controlled macromolecules, called dendrimers [1], has provided a chemical realization for the abstract construct of Bethe lattices (Cayley trees), which have been used before as a "toy model" for a range of problems, such as diffusion on non-euclidean lattices [2]. Energy transfer and funneling on antenna-like dendrimer supermolecules have been investigated recently, as well as the underlying quantum mechanics of excitation localization and delocalization [3–5]. As expected it was found that these properties strongly depend on the particular geometry of the prepared molecule. For example, for a class of dendrimers made of purely hydrocarbon chains, which is composed of repeated phenylacetylene (PA) units, two series of dendrimers have been examined in the past, the "compact" series with ideal fractal (dilation) symmetry, in which all dendrimeric subunits are composed of identical single PA chains, and the "extended" series which have PA linear chains that increase in length toward the molecular locus. These PA dendrimers and also their perylenic-substituted derivatives have been studied both theoretically [5,6] and experimentally [4,7,8] with respect to the energetic absorption and funneling characteristics. In both cases, due to the *meta*-position branching geometry at each node, these dendrimers exhibit excited state localization along individual PA

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chain units [3,7]. Thus, it is possible to observe exciton hopping between states. In addition, it has been shown previously [4] that due to the subtle change in molecular geometry between the compacts and extendeds, these two groups display markedly different energetic behavior. In particular, the extended dendrimers, with the inherent thermodynamic bias for exciton trapping at the molecular locus, show greatly enhanced energy funneling capabilities, with nearly unit quantum yield of exciton transfer from the dendrimer backbone to a perylenic trap substituted at the molecular core [4]. In contrast, the compacts exhibit a bias for exciton hopping outward, toward the dendrimer periphery (rather than toward the locus), resulting in greatly decreased rates and efficiencies of exciton trapping. This funneling behavior has been observed to vastly increase the trapping photoefficiency in several supermolecular systems [9,10], and may provide a useful means for interconversion of electrical, optical, and chemical energy.

As excitation localization often leads to exciton hopping and/or fusion, connections can easily be made with stochastic models [11–18] that involve well known ideas from random walks. One frequently used quantity in this area that may be pertinent here and is related to diffusion has been S_N, "the average number of distinct lattice sites visited by a simple random walker after N steps" [2,13,16–18]. Another quantity has been the survival probability of an ensemble of random walkers undergoing an elementary bimolecular reaction [11,12,14,15]. The reaction can be the "one kind" $A + A \rightarrow$ products reaction [14,19] or the "two kind" $A + B \rightarrow$ products reaction [11,12,15]. Classically, for $A + A$, and for equimolar $A + B$, the survival probability (density $\rho = \rho_A = \rho_B$) is given by:

$$1/\rho - 1/\rho_0 = kt \tag{1}$$

where ρ_0 is the density at $t = 0$ and k is the "rate constant." Asymptotically (for $t \rightarrow \infty$), this gives

$$\rho^{-1} = kt \tag{2}$$

In non-classical kinetics [11–15,20] this relation is generalized to

$$
\begin{aligned}
&\rho^{-1} = kt^f, \quad 1/2 \leqslant f \leqslant 1 \\
&f = d_s/2, \quad d_s \leqslant 2 \\
&f = 1, \quad d_s > 2
\end{aligned}
\tag{3a}
$$

for the $A + A$ reaction and to

$$
\begin{aligned}
&\rho^{-1} = kt^f, \quad 1/4 \leqslant f \leqslant 1 \\
&f = d_s/4, \quad d_s \leqslant 4 \\
&f = 1, \quad d_s > 4
\end{aligned}
\tag{3b}
$$

for the $A + B$ reaction, and where d_s is the spectral dimension.

Similar to Ref. [13], and using the symbol S_t instead of S_N,

$$S_t \sim t^f, \quad 1/2 \leqslant f \leqslant 1 \tag{4}$$

where $f = d_s/2$. However, for infinitely extended Bethe lattices (irrespective of the dimension of the embedding euclidean space) the asymptotic expression [4] has $f = 1$ (because $d_s > 2$) and is explicitly [2]:

$$S_N = \frac{z-2}{z-1} N, \quad N \rightarrow \infty \tag{5}$$

where z is the coordination number of the site, i.e. the number of bonds emanating from each site. Obviously N is just a discrete (integer) expression of the time t.

In treating this quantity there are two factors in the dendrimer structures that are inherently different from regular lattices or other customary molecular systems. The first is the property that a regular random walk on a dendrimer is in effect a "biased" walk with the probability of hops towards the periphery highly enhanced over that towards the center. This is simply because there are two or more paths in that direction as opposed to one path in the opposite direction. An analogous situation occurs in charge transfer calculations in the *para* and the *meta* positions of ring structures [26–28], where the efficiency of the process varies significantly with the structure. The second factor, related to the first, is that the boundaries of the structures are reached fairly fast, and one should be aware of the presence of finite-size effects, even at early times, and for large systems.

One point of interest in this work is to test, via Monte Carlo simulations [13–18], the validity of Eq. (5) for finite lattices and finite times. The other points of interest involve the A + A and A + B reactions. Do they obey the classical expression (Eq. (2)) or the non-classical equation (3)? Also, does the scaling relation for the A + A and A + B reactions [20],

$$\rho^{-1} \sim S_t \qquad \text{for A + A} \tag{6}$$

$$\rho^{-1} \sim S_t^{1/2} \quad \text{for A + B} \ d \leqslant 2 \tag{7}$$

which seems to hold for both euclidean and fractal lattices, still hold for Bethe lattices, i.e. dendrimers? Somewhat unexpectedly the answer to this question is negative, even though the scaling given by Eq. (5) is preserved for finite times and sizes. We note that for the very distinct problem of immobile reactants [29] the solutions are entirely different.

2. Monte Carlo simulations

We construct dendrimer structures with the coordination number, z, and the generation order, g, being parameters of the system [1–3]. We also use k, the number of emanating branches, where $z = k + 1$. A schematic of some typical structures is given in Fig. 1, for $z = 3$, 4, and 5. Typically, g can go up to 22 (about 8 million sites for $z = 3$), while z takes the values of $z = 3$, 4, or 5. Due to the peculiarity of the dendrimer geometry, no underlying lattice can be used here, but instead one uses an indexing mechanism. On a given node with a given index, this mechanism relates, by use of simple algebraic relations, the current index value to the indices of all neighboring sites, and one must only be careful to keep the correct indexing of all nodes. A particle is placed at a random position (node) on the structure, and it is allowed to diffuse, by performing a random walk, to the neighboring nodes only, one node at a time (one time unit, or one Monte Carlo step). As shown later, the boundary conditions in this problem play a vital role, and thus we have used various boundary conditions. While, in general, all nodes have equal probability for the transfer, by its nature the structure is such that particles move with higher probability towards the periphery, rather than the center of the molecule [6]. This probability depends, of course, on z. The higher the z value, the higher the rate of transfer to the periphery. For a single particle we monitor the number of nodes that are visited during the random walk, as a function of time. For interacting (reacting) particles, the simulation is performed similarly to the case where we use a lattice, but with the lattice points now being the branching nodes of the molecule. Simulation details for the reaction mechanism have been frequently reported in the past [21]. A large number
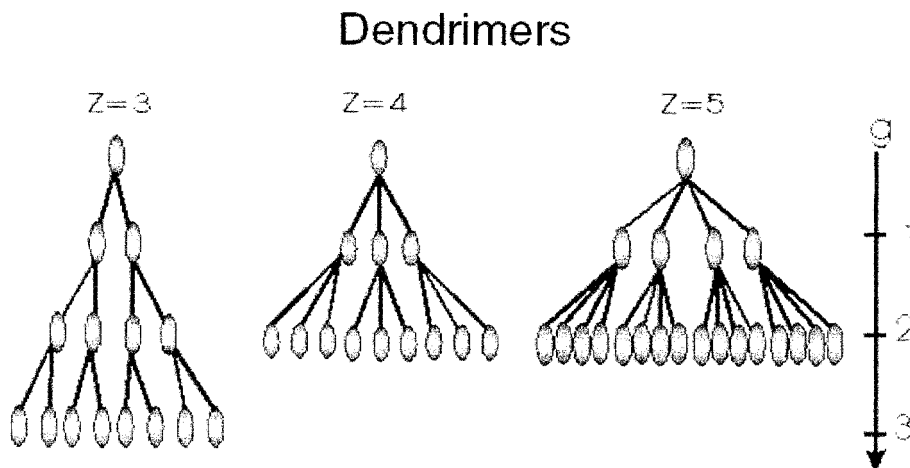
Dendrimers

Fig. 1. Schematic of three dendrimers, with $z = 3$, 4, and 5. The generation order, g, is as shown, $g = 3$ (for $z = 3$), and $g = 2$ (for $z = 4$ and 5).

of particles occupy the structure's nodes, perform random walks, and upon encounter interact by annihilation. Here, we monitor the population of particles, as a function of time.

3. Exploration space S_N

This is the number of distinct sites visited at least once in N steps. The theoretically expected value for S_N for Bethe lattices (Cayley trees) is given [2] by Eq. (5). This is an asymptotic behavior for very large values of N. Our calculations are given in Figs. 2–4. We note that there are several parameters that affect these curves, which do not appear in the asymptotic formula, as these are all due to finite-size effects. In Fig. 2, for $g = 20$ and $z = 3$, we have a different curve depending on the origin of the random walk, the origin being (a) the core of the dendrimer, $x = 0$, (b) the middle of one of the branches, randomly chosen, which here is at the 10th generation, and (c) any point randomly chosen on the dendrimer. The asymptotic formula (Eq. (5)) is also shown, where, for $z = 3$, we have simply that $S_N = N/2$. We note (as seen below) that in the range of $N = 100$ steps the calculations
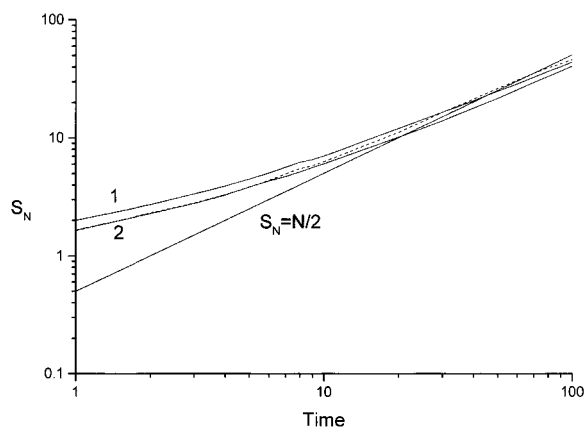

Fig. 2. S_N as a function of time, for random walks on dendrimer structures of generation order $g = 20$, and coordination number $z = 3$, for different origins of the particle: (1) randomly chosen site, (2) the core of the dendrimer, and (3, dotted line) a point on the middle generation, at $g = 10$. Also shown is the asymptotically expected $N/2$ straight line. Reflective boundary conditions are used here.
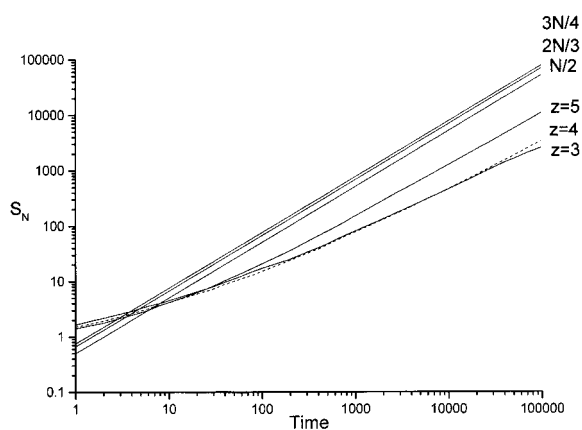

Fig. 3. S_N as a function of time, for random walks on dendrimer structures of generation order $g = 11$, for different z values, $z = 3$, 4, and 5. The $N/2$, $2N/3$, and $3N/4$ lines, which are the asymptotic solutions (Eq. (5)), are also drawn. Reflective boundary conditions are used here.

are close to the theoretically predicted value, but for larger N the calculated S_N values are always smaller.

In Fig. 3 we show the effect of the coordination number z, for $z = 3$, 4, and 5. The larger the z, the larger the values of S_N, as expected. Here the calculations are for $g = 11$, but the trees with larger z obviously contain a larger number of sites. The use of an identical g assumes that the mean time to the periphery is of the same order for all cases [6], even though there are some small differences (faster rate with increasing z). Again the absolute S_N results are quite smaller than the theoretically predicted values. We note that it is practically impossible to recover the asymptotic formula with a simulation calculation, because even though the structures contain several million sites, nevertheless, the boundaries are reached very quickly (order of 100 steps or less), due to the nature of the dendrimer structure (where periodic boundary conditions cannot be applied). This is in contrast to S_N on euclidean and fractal lattices. Thus, one realizes that in practicality this problem is a problem of finite-size structures rather than given by a theoretical model based on infinite extension. Increasing the size of the dendrimer structures by several more generations would not affect this situation, and one would need values of g in the tens or
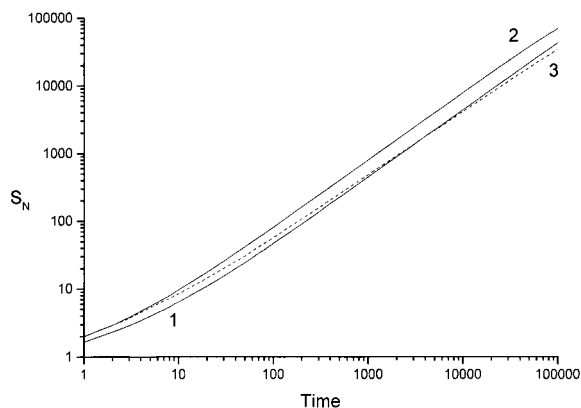
Fig. 4. S_N as a function of time, for random walks on dendrimer structures of generation order $g = 22$, and coordination number $z = 3$, for different sets of boundary conditions: (1) reflective boundary conditions, (2) cyclic conditions, where the particle reaching the periphery of the dendrimer is positioned on the core and vice versa, and (3, dotted line) cyclic conditions, where the particle reaching the periphery is positioned randomly on any other site of the structure.

hundreds of thousands to approach the asymptotic result, something that is prohibitively expensive for simulations and irrelevant to experiments.

In Fig. 4 we employ some types of periodic conditions, even though the nature of the structures makes it rather arbitrary. We also compare with the case of non-periodic conditions (reflective boundary conditions). In particular, we use two models: In the first model, when the particle reaches the periphery of the structure it is repositioned on the $x = 0$ site (the core site). When the particle moves from the core site to a lowered index site, then it is repositioned on a randomly chosen structure site. In the second, when the particle reaches the periphery of the structure it is repositioned on another structure site randomly chosen. The numerical results for generation order $g = 22$ and for $N = 100\,000$ steps show a ratio of $S_N/N = 0.43$ for the case of reflective boundary conditions. They give a result of 0.70 for the first model, and 0.35 for the second model. The asymptotically expected result is 0.50. We observe large variations, including the case where the result is larger than that theoretically predicted. This happens for the first model above, where the repositioning on a new site results in placing the

particle in a new and unexplored region, which may have little to do with reality. We note that the calculated exponent f of Eq. (4) is close to unity, so the scaling agrees with Eq. (5). We finally note that the S_N numerical values in Figs. 3 and 4 may seem contradictory, but in view of the above we clearly understand that this is due to the different g values used. The $g = 11$ and the $g = 22$ cases vary tremendously in size. The $g = 11$ contains about 4000 molecules, the $g = 22$ contains 8 million molecules (for $z = 3$). It is thus expected that huge differences in the S_N behavior will be observed, all due to finite-size effects.

4. A + A and A + B reactions

In Fig. 5 we show the quantity $1/\rho - 1/\rho_0$ for the A + A reaction on a dendrimer, for three different structures, (a) $g = 20$, $z = 3$, (b) $g = 13$, $z = 4$, and (c) $g = 10$, $z = 5$ (chosen to roughly preserve the total number of sites). We observe that this quantity scales with time, with an exponent of $f = 0.80 \pm 0.03$. In Fig. 6 we show the same calculation for the A + B model, and here the scaling exponent has the value of $f = 0.50 \pm 0.03$. Also shown here is the effect of the coordination number, where we see that this effect
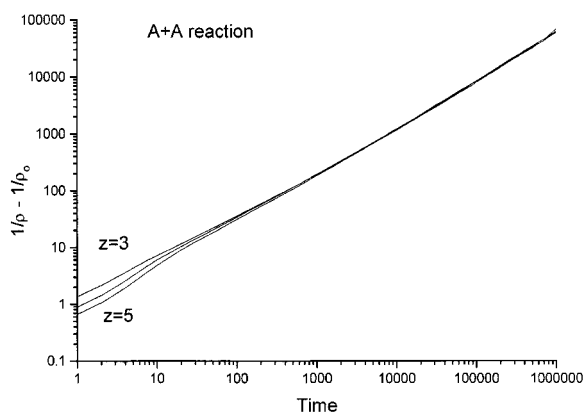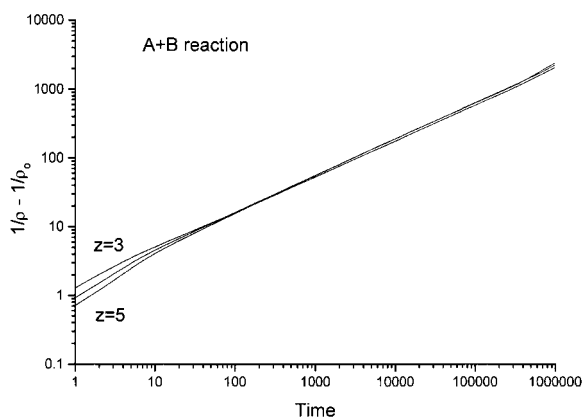

Fig. 5. Plot of the reaction progress $1/\rho - 1/\rho_0$ as a function of time, for the A + A = 0 reaction on a dendrimer, for three different generation orders and corresponding coordination numbers, as follows: ($z = 3$, $g = 20$, 2 097 151 sites), ($z = 4$, $g = 13$, 2 391 484 sites), and ($z = 5$, $g = 10$, 1 398 101 sites). The initial density was $\rho_0 = 0.8$.

Fig. 6. Plot of the reaction progress $1/\rho - 1/\rho_0$ as a function of time, for the $A + B = 0$ reaction on a dendrimer, for three different generation orders and corresponding coordination numbers, as follows: ($z = 3$, $g = 20$), ($z = 4$, $g = 13$), and ($z = 5$, $g = 10$). The initial density was $\rho_0 = 0.4$ per species.

is only important at early times, while after $t = 100$ steps there is practically no difference due to z. These calculated f values are significantly different from unity. While we have a very good and detailed knowledge for f and for the scaling of reactions in other lattices [22,23], fractals [24], anisotropic systems (tubes, pores, etc.) [25], the present case of dendrimers (Bethe lattices) does not fall in any of these categories, presumably due to the inherently different nature of the dimensionality and/or topology, and thus we have no ground for comparison. Additionally, we note that while for the random walk of an individual particle the finite-size effects predominate in the numerical calculations (as discussed above), here this is not expected, since the particle–particle annihilations are the determining factor, and should prevent any particle from sampling the entire lattice.

5. Conclusions

We have investigated numerically the dynamics of random walks on dendrimer model systems. We evaluated S_N, for individual particles, and the rate of annihilation for multiple reacting particles on dendrimers. For S_N, the finite structure, finite time, simulations do not give the Hughes–Sahimi asy-

mptotic result, but the discrepancy is only in the prefactor ($z - 2/z - 1$). However, the linear character of the scaling is preserved in these finite systems, i.e. $S_N \sim N$.

On the other hand, both the $A + A$ and the $A + B$ reactions do not give the classical scaling, i.e. ($1/\rho - 1/\rho_0$) is not linear in time. This is particularly surprising since the S_N relation is linear in time. Thus, the usual scaling, obeyed for both classical reactions and for low dimensional and fractal reactions (Eqs. (3) and (6)), does not appear to be valid here. The reasons for that and for the specific values of the scaling exponents are not clear at this point. However, we find (see Appendix A) an analogous occurrence for the biased walk in one-dimension, a case that is not related topologically, but nevertheless involves the characteristics of the biased walk (preferred direction of motion).

Our results are in general qualitative agreement with the recent work of Bar-Haim and Klafter, who investigated the possibility of using dendrimers as light harvesting antennae by probing the geometric vs. energetic competition within the structure [6,30–32].

Acknowledgements

Appendix A. Biased walk on one-dimensional lattices

Hopping on the Bethe lattice has been mapped [6] onto the problem of biased walk on a one-dimensional lattice, for the mean passage time, where this is an exact mapping. However, this is not the case for S_N, or for the reaction problems. These problems have been studied in detail before [33–40], nevertheless, in order to see the numerical analogy to the dendrimer case we performed simulations for S_N, and for the reactions $A + A$ and $A + B$ on a one-dimensional lattice with a preferred direction designated by a probability $p > 1/2$. The case of $p = 0.5$ is the case of an un-
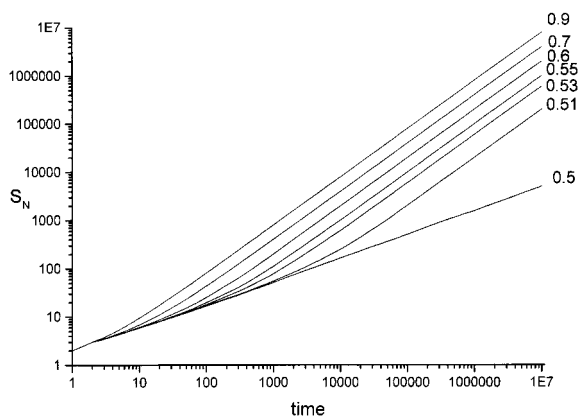
Fig. 7. Plot of S_N vs. time, for a biased random walk, in one-dimensional lattices, as given by the bias parameter p, for the cases of $p = 0.5$ (no bias at all), and $p = 0.51, 0.53, 0.55, 0.6, 0.7$, and 0.9 (bottom to top in the figure). The lattice size was 1 million sites, and a total of 500 realizations were averaged for this data.
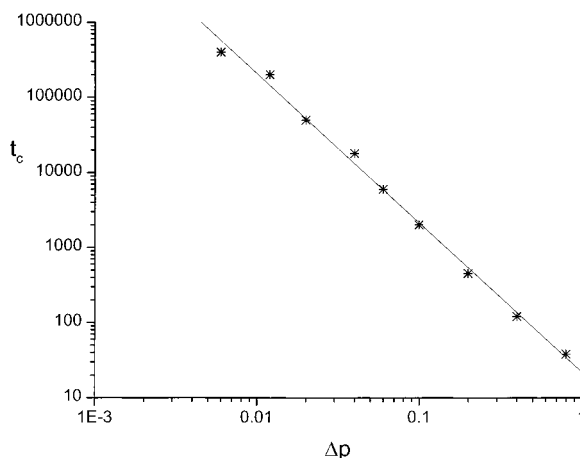

Fig. 8. The crossover time, t_c, as a function of Δp for the data of Fig. 7. The slope is, as expected, -1.98 ± 0.05. The points are the calculated data, while the straight line is the best fit.

biased (fully stochastic random) walk, while the case of $p = 1$ is the case of fully ballistic motion. We used p values in the range $p = 0.51$–0.90. Fig. 7 shows the results for the S_N calculations, where we see that the $p = 0.5$ case has a slope of $1/2$, as expected, while as p is slightly increased, even just to $p = 0.51$, there is a drastic change to a slope of 1, which is the case for all $p > 0.5$ values. The change from the slope of $1/2$ to the slope of 1 occurs with a crossover time t_c. The particles move with a drift velocity v, and the crossover time happens when vt is equal to the distance R traversed during the walk. Thus

$$vt = R \tag{A.1}$$

From the diffusion equation $R^2 = 2Dt$. Also, v is proportional to Δp, where Δp is the difference between the drift to the right and that to the left, i.e. $\Delta p = p - (1 - p) = 2p - 1$. Therefore,

$$(\Delta p)t \sim (2Dt)^{1/2} \tag{A.2}$$

leading to:

$$t_c \sim 2D/(\Delta p)^2 \tag{A.3}$$

Indeed in Fig. 8 we plot the crossover time t_c vs. Δp, and we see that the slope is approximately equal to -2, verifying the crossover conjecture.

Finally, under cyclic boundary conditions we find that the exponent f is $1/2$ for the $A + A$ reactions and $1/4$ for the $A + B$ reactions (see Fig. 9). This is exactly the same result as for these reactions with ordinary, unbiased random walk [14,15]. In Fig. 9 we show three different values of p, for both models, where this result is clear. The kinetic model employed for all reactions is that of
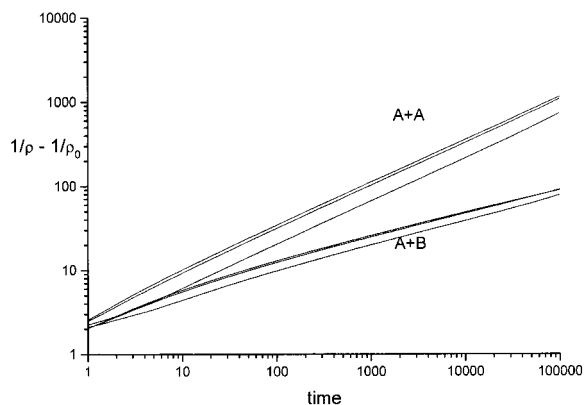

Fig. 9. Plot of the reaction progress $1/\rho - 1/\rho_0$ as a function of time, for the $A + A = 0$, and for the $A + B = 0$ reactions, for three values of the parameter p, $p = 0.5, 0.7$, and 0.9, bottom to top, for both cases. The lattice size was 1 million sites, and a total of 25 realizations were used. The initial density was $\rho_0 = 0.8$ (for the $A + A$ reaction), and $\rho_0 = 0.4$ (for the $A + B$ reaction).

excluded volume. We believe that this is an interesting case of scaling breakdown, especially for the A + A reaction, for which we are not aware of any precedents in euclidean, fractal or anisotropic lattices. We also note that our $p < 1$ results differ from the asymptotic exponent of $f = 1/3$ for the A + B reaction [34–40]. However, only for the ("unphysical") $p = 1$ case have literature simulations [34,35] given $f = 0.30$. Apparently much longer time simulations may be required to reach this limit for $p < 1$. On the other hand, our A + A results are in full agreement with the $p = 1$ simulations and conclusions of Privman et al. [41], i.e. in this case the universality class is the same for biased and unbiased walks (in contrast to S_N and possibly the A + B case).

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