Energy Transport and Trapping in Polymeric Media: Small-World Networks†

A. Blumen* and F. Jasch‡

Theoretische Polymerphysik, Universität Freiburg, Hermann-Herder-Strasse 3, D-79104 Freiburg, Germany

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Energy transfer in polymers is characterized by nonexponential decays, mostly due to the disorder of the underlying medium. In this work we take as models for the medium small-world networks (SWNs). SWNs are built starting from regular lattices (say from linear chains) through the insertion (with probability p) of additional links, which then connect distant pairs of sites. In this way SWNs combine random and regular features. As a dynamical problem we evaluate the energy migration followed by trapping (quenching) by acceptors, randomly distributed over the SWN, and compare the trapping decay to the forms found when the underlying structures are regular lattices, fractals or ultrametric spaces; as we show, trapping on SWNs displays new decay aspects.

1. Introduction

Energy transport and trapping in polymeric media is a subject of continuous interest. When the incoherent transport dominates, one often describes the motion of the excitations over the donors through random walks (RW).[1−3] A process of much interest is quenching,[4−6] by which the excitation leaves the donor subsystem by being transferred to an acceptor; in the RW-picture the process is termed trapping[1,2] and it corresponds[2,3,7] to the reaction $A + B \rightarrow B$. As is well-known by now, even when the density of excitations is very low, their decay is seldom exponential; trapping is characterized by typical departures from exponentiality, which depend on the dimensionality of the lattice over which the walk takes place and on the disorder (geometric, temporal or energetic).[1,2,3,7] Much work has focused on trapping on regular lattices,[1,2,8] on percolating and on fractal networks,[3,9] and also on cases which involve energetic disorder; here descriptions in terms of ultrametric spaces (UMS) have turned out to be very fruitful.[10−12] In this work we extend such random-walk approaches to small world networks (SWNs).[13−18] Small-world networks (SWN) are a special class of random graphs;[19] they provide an elegant way to model the randomness encountered in systems which are neither fully random nor fully regular;[13−24] in this way they complement previous approaches based either on fractals,[3,9,25,26] which depict situations determined by geometric disorder (such as percolation), or based on UMS,[10−12] which focus on the energetic aspects of randomness. Experimentally, energy transfer in such complex media gets to be more and more accessible, due to advances in modern fluorescence techniques and in the fixing of chromophores to macromolecular systems.[27−29]

The paper is structured as follows: In section 2 we introduce the concept of SWNs and show that it allows to interpolate between fully regular and fully random networks. Section 3 is devoted to the trapping problem on SWNs. Now trapping is very sensitive to disorder; leaving aside the aspects related with the "Lifshitz-tails", which give rise to long-time decay-forms associated with rare events,[1,2,30−35] we will focus here on the short and medium-time decay patterns, where cumulant expansions[8] allow to approximate fairly well the part of the decay which is readily accessible to experiments. In section 4 we contrast these results to the decay forms which obtain on low-dimensional systems, on fractal structures, and on UMS.

2. Small-World Networks (SWN)

In disordered media, such as polymeric materials, one is often confronted with coexisting regular and stochastic features. This finding is quite general: Neglecting the causes leading to the networks' creation, and focusing on the emerging structures only, one finds that things as diverse as air-traffic networks, electrical power grids, neural nets and polymer networks have in common that they are neither fully regular nor completely random.[13−15,36] One also observes that in such networks the minimal (chemical) distance between any two sites scales logarithmically (as in random graphs) with the networks' size.[14,19,36] Now a procedure to model such networks was introduced in ref 13: The starting point is a regular lattice, with bonds connecting all sites whose mutual distance is less than a preassigned, small length. Then a small number of additional links (ALs), of arbitrary length, are added to the network. Such networks are then termed small world networks (SWNs). Interestingly, even a very small density of ALs changes drastically the network's properties, such as given by its vibrational density of states,[17] its stretching by external means,[22,23] or the diffusion of particles over it.[20,21]

One is naturally led to SWN-ideas when considering energy transport over polymer chains; there, realistically, the excitation's motion is not restricted only to steps along the backbone but (due to the fact that monomers far away along the backbone may be close to each other in space), also energy transfer between such monomer-pairs is possible.[37−41] In fact several experimental groups report that in one-dimensional (1D) systems the dynamics of the backbone facilitates departures from the simple picture of transport along linear chains.[42] This suggests enlarging the model by the introduction of ALs between pairs of monomers.

In our case we choose as underlying regular lattice for the SWN a ring with L sites (vertexes), where $L \gg 1$, each site being connected to its two nearest neighbors (NN). Then we add to each site an AL, with probability p. The other end of the

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* To whom correspondence should be addressed. Fax-Number: 49-761-203-5906. E-mail: blumen@physik.uni-freiburg.de.

‡ E-mail: florian@physik.uni-freiburg.de.

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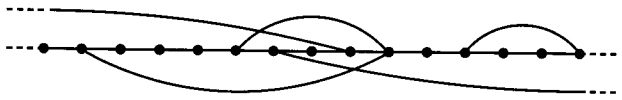

Figure 1. Sketch of part of a SWN, built from a ring. Some ALs connect to sites outside the figure.

AL gets attached, with equal probability, to any of the other sites. In this way we add on the average *pL* new bonds (ALs) to the ring. Figure 1 displays a small portion of such a SWN-realization. We hasten to note that (as shown in recent works) SWN with ALs attached corresponding to a scaling law,[21] on one hand, and self-avoiding polymer chains (SAW) with ALs added in places where two monomers get together,[37−39] on the other, may differ in their dynamical behavior.[43]

While the first works on SWNs focused mainly on their static (geometric) properties, nowadays dynamical aspects start to gain importance; among the subjects treated recently we mention the dynamics of Ising models on SWNs,[16] the spectral properties of SWN-Laplacians,[17] the spreading of diseases on SWNs,[44] and problems related to chemical physics, such as the target problem on SWNs[18] and the modeling of polymers' dynamics by SWN.[22,23] In the following we will examine the trapping of electronic excitations in media described by SWNs.

3. The Trapping Problem

We start to model the trapping of electronic excitations in terms of the A + B → B reaction, and take the initial number A_0 of excitations to be much less than the number of quenchers B, i.e., $A_0 \ll B$. In this case, under well-stirred conditions, chemical kinetics predicts an exponential decay law:[2]

$$A(t) \simeq A_0 \mathrm{e}^{-Bkt} \tag{1}$$

In this section we focus on deviations from the exponential behavior of eq 1 due to the fact that the Bs are stationary (traps). As mentioned above, the trapping problem (in which the A particles move and get annihilated by stationary B traps) has received considerable attention due to its marked departures from exponentiality at very long times:[30−35,45] this aspect is due to the statistics of rare events and it is still of much current interest.[34,35,46] We do not consider these aspects here, since we want to focus on the experimentally accessible region, which also shows clear departures from exponentiality.[8,45]

As stated in the previous section, we model the dynamics by letting the excitation move randomly over the sites of the underlying structure. In former works we studied such walks on regular and on fractal lattices.[2,7,47−49] For any given lattice stepwise motion is a Markov process, defined by specifying all the transition probabilities w_{fi} of going from site i to site f in one step. If one stipulates that at each time step the particle has to leave the site it just occupies and if all neighbors are equally prone to receive it, then $w_{fi} = 1/z_i$, where z_i is the connectivity of site i. The master equation for the probabilities $P_i(n)$ of being at site i after the nth step reads:

$$P_i(n+1) - P_i(n) = \sum_{j=1}^{N} w_{ij} P_j(n) - P_i(n) \sum_{j=1}^{N} w_{ji} \tag{2}$$

where the i and j denote the sites of the underlying lattice. Equation 2 is nothing but the discrete-time variant of diffusion on the given lattice.

Let us focus on what happens in the presence of traps. Now trapping is considerably more difficult to study than $P_i(n)$, since for it no expression like eq 2 is known in general, and one has to use numerical simulation procedures. Previously, much work has focused on diffusion and trapping on regular lattices,[1,2,50−52] on fractals (see, for example. refs 2, 9, 45, 48 and references therein), on Cayley trees,[53−55] on dendrimers,[56] and on UMS.[10−12] Here we follow our study of RWs on SWNs,[18,20,21] while focusing on trapping.

For trapping numerical simulations are the method of choice; in these a walker performs at fixed time intervals a step to one of its NN (for the SWNs considered here either along the ring or over an AL); trapping occurs instantaneously, if the walker lands on a site occupied by a B. For a particular realization of the random walk of the excitation let R_n denote the number of distinct sites visited by it in the first n steps (usually one sets $R_0 \equiv 1$). For the same realization of the walk let F_n denote the probability that trapping has not occurred up to the nth step. If the traps are placed randomly over the lattice, with probability q, one has[8]

$$F_n = (1-q)^{R_n - 1} = \mathrm{e}^{-\gamma(R_n - 1)} \tag{3}$$

where we have set $(1-q) \equiv \mathrm{e}^{-\gamma}$ and assumed the origin of the walk not to be a trap. The measurable survival probability is the average of F_n over all the realizations of RWs and, in our case here, also of the underlying SWNs; therefore,

$$\tilde{\Phi}_n = \langle F_n \rangle = \mathrm{e}^{\gamma} \langle \mathrm{e}^{-\gamma R_n} \rangle \equiv e^{\gamma} \Phi_n \tag{4}$$

In the following we focus on Φ_n, which is an average over an exponential expession: a powerful way to proceed is to express it as an exponent of averaged quantities, the so-called cumulants.[8] In this way, one is lead to

$$\Phi_n = \langle \mathrm{e}^{-\gamma R_n} \rangle = \exp[\sum_{j=1}^{\infty} K_{j,n} (-\gamma)^j / j!] \tag{5}$$

where the $K_{j,n}$ are the semiinvariants (cumulants) of the distribution of R_n, i.e., $K_{1,n} = \langle R_n \rangle \equiv S_n$, $K_{2,n} = \langle R_n^2 \rangle - \langle R_n \rangle^2 \equiv \sigma_n^2$, etc. As discussed in ref 8, we can now truncate the sum on the right-hand side of eq 5:

$$\Phi_{i,n} = \exp[\sum_{j=1}^{i} K_{j,n} (-\gamma)^j / j!] \tag{6}$$

and use the $\Phi_{i,n}$ as increasingly reliable approximations for Φ_n at short and medium times (i.e., n). On a note of caution we note that this procedure does not capture the asymptotics of Φ_n, given that for $n \to \infty$ in general Φ_n is not an analytical function of γ anymore.

Exemplarily, the simplest nontrivial decay form is

$$\Phi_{1,n} = \exp(-\gamma S_n) \tag{7}$$

Now $S_n \equiv \langle R_n \rangle$, the mean number of distinct lattice sites visited in n-steps, has a long history of study. Montroll and Weiss have shown that it can be calculated for regular d-dimensional lattices using generating function techniques.[1,50−52] One finds, depending on the dimension and for not too small n:[51,52]

$$S_n \sim n^{1/2} \qquad (d = 1) \tag{8}$$

$$S_n \sim n/\ln(n) \qquad (d = 2) \tag{9}$$

and

$$S_n \sim n \qquad (d = 3) \tag{10}$$

For fractals, S_n depends on the spectral dimension $\tilde{d}$; it obeys:[57]

$$S_n \sim n^{\tilde{d}/2} \qquad (\text{for } \tilde{d} < 2) \tag{11}$$

and follows eq 10 for $\tilde{d} > 2$. Hence $\tilde{d} = 2$ is the marginal dimension for the mean number of distinct sites visited. A similar situation occurs for UMS, where[10]

$$S_n \sim n^{\mu} \qquad (\text{for } \mu < 1) \tag{12}$$

where $\mu = (\ln b)k_BT/\Delta$, with $b + 1$ being the connectivity and $\exp(-k_BT/\Delta)$ the Boltzmann factor for thermally activated steps. On the other hand, for $\mu > 1$ one recovers eq 10. The same holds true for RW over nontrivial (*i.e.* $b > 1$) Cayley trees. In all these cases, evidently, by inserting eqs 8, 9, 11, and 12 into eq 7 one is led to nonexponential decay patterns.

Given that the domain of validity of $\Phi_{1,n}$ may be quite restricted (as for instance in $d = 1$, eq 8) one may now try to go stepwise higher in the approximation.

Thus including the variance, σ_n^2, leads to

$$\Phi_{2,n} = \exp(-\gamma S_n + \gamma^2 \sigma_n^2/2) \tag{13}$$

Further terms of the cumulant expansion eq 6 may be determined from the higher moments of R_n. We note, however, that even for regular lattices the determination of the higher cumulants of R_n is an arduous task.

As a first approximation for evaluating trapping on SWN, we focus now on $\Phi_{1,n}^{SWN} = \exp(-\gamma S_n^{SWN})$, and hence on S_n^{SWN}. As shown in ref 18 through numerical simulations, S_n^{SWN} is very well described by the following scaling relation:

$$S_n^{SWN} = n^{1/2} f(np^2) \tag{14}$$

where $f(x) = \sqrt{8/\pi}$ for $x \to 0$ and $f(x) = C\sqrt{x}$ for $x \to \infty$. Note that for very small n or p, such as $np^2 \ll 1$, one recovers the one-dimensional character of S_n, eq 8. This was to be expected, since, for a small number of steps and very few ALs, a walker sees mainly 1D surroundings. On the other hand, for n large the walker explores a quite open structure, not very different from a random tree.[46] Hence, on SWNs, one has a temporal transition with respect to S_n between a confined situation at small n and an open one for n large. Evidently, eq 7 again leads to a nonexponential behavior for small n. For n large Φ_n stays nonexponential, due to the higher $\Phi_{i,n}$ forms.

Given that even S_n^{SWN} is known (at least for the moment) only numerically, we focus now on the numerical evaluation of Φ_n. We note from the start that placing the traps randomly over different SWNs and simulating walks which stop on trapping would be an extremely computer time-consuming procedure. We prefer instead to work using eq 5, since it offers the great advantage of being, in fact, a q-independent algorithm: as is evident from eq 5, q appears in it only through γ, a parameter. Numerically one has only to determine the R_n values on SWN-lattices *devoid* of traps, evidently, however, for many SWN-realizations.

We start our procedure as follows: For a given q we construct 10 different SWNs, of size $L = 9 \times 10^5$ each. On these we simulate a total of 10^5 RWs with randomly chosen starting points, and determine the R_n for each walk; from these we obtain the R_n-distribution. By choosing the starting points randomly we sample, in fact, a very large class of local SWN-geometries, much larger than what the 10 SWN-realizations indicate at first glance. The so-determined R_n-distribution allows us then, via eqs 5 and 6, to evaluate numerically (for arbitrary q-values) both Φ_n^{SWN} and also the corresponding $\Phi_{i,n}^{SWN}$. The results of these calculations are presented in Figures 3 and 4, whereas Figure 2 shows trapping on the 1D chain.

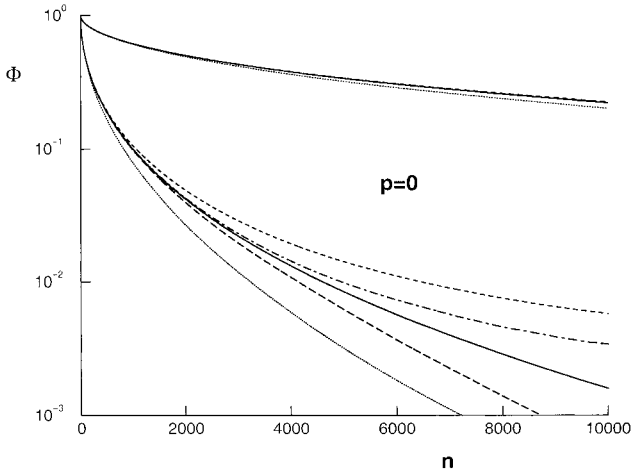

Figure 2. Decay due to trapping on a linear chain devoid of ALs ($p = 0$); the trap densities are $q = 0.01$ for the upper and $q = 0.05$ for the lower curves. The full lines give Φ_n and the other lines $\Phi_{1,n}$ to $\Phi_{4,n}$. The approximations are $\Phi_{1,n}$ (dotted), $\Phi_{2,n}$ (short-dashed), $\Phi_{3,n}$ (long-dashed), and $\Phi_{4,n}$ (dot−dash).

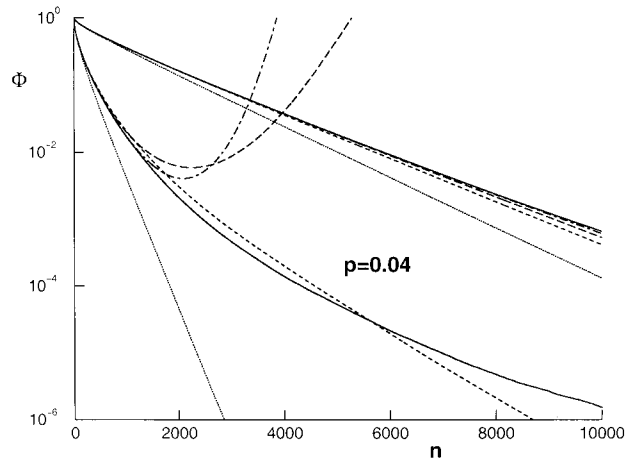

Figure 3. Survival probabilities Φ_n (solid lines) on SWNs with an AL-density of $p = 0.04$, compared to the corresponding cumulant approximations $\Phi_{1,n}$ (dotted), $\Phi_{2,n}$ (short-dashed), $\Phi_{3,n}$ (long-dashed), and $\Phi_{4,n}$ (dot−dash). The trap desities are $q = 0.01$ for the upper and $q = 0.05$ for the lower curves.

In Figure 2 we depict trapping on a linear chain. Note that this corresponds faithfully to our SWN-model (a ring) for $p = 0$, since we keep the number of steps below $n = 10000$, and hence the limitations of the ring cannot be seen. Displayed are in logarithmic scales the decays for the trap densities $q = 0.01$ and $q = 0.05$. We present both the exact decay forms and also the first four approximations, as given by eq 6. The results reproduce those of ref 8. Clearly evident from Figure 2 is the nonexponential character of the decay. Furthermore, while $\Phi_{1,n}$ provides a reasonable description for the decay in the first decade, it already fails to provide it in the second and further decades. In the second decade, visible in the Figure only for $q = 0.05$, the form $\Phi_{4,n}$ turns out to be quite reasonable. One may also note the fact that the increasing cumulants approximate the true decay from below (if they are odd) and from above (if they are even); this is due to the fact that all four cumulants considered are strictly positive, and to the changes of sign in the exponent of eq 6.

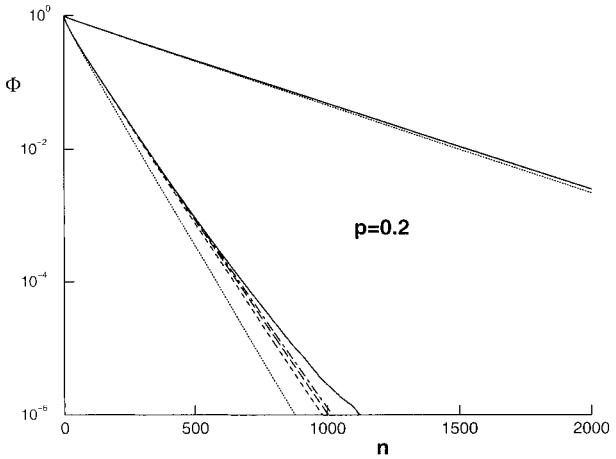

Figure 4. The same as in Figure 3 for SWNs with an AL-density of $p = 0.2$. In the upper group of curves (for $q = 0.01$) in the range of the figure only $\Phi_{1,n}$ can be distinguished from the true decay Φ_n.

The situation changes when we go to nontrivial ($p \neq 0$) SWNs. In Figures 3 and 4 we display the trapping decay for concentrations of ALs equal to $p = 0.04$ and to $p = 0.2$, respectively. One expects with increasing p that the SWN will behave in a more and more regular manner, at least in what the first few decades of the decay are concerned. This is also the case when one looks at the decay for $q = 0.01$; there, already for $p = 0.2$, the description in terms of $\Phi_{1,n}$ is very satisfactory for the first two decades shown; in fact, the decay itself is hardly distinguishable from an exponential. Also the situation for $p = 0.2$ and $q = 0.05$ is satisfactory; as can be inferred from Figure 2, $\Phi_{1,n}$ is not bad at all over the first two decades of the decay, and the description which uses $\Phi_{2,n}$ performs very nicely over 4 orders of magnitude in the decay (possibly the experimental limit nowadays), a region in which nonexponential features appear quite clearly. In the intermediate region, for $p = 0.04$, the situation is more complex, as displayed in Figure 3. Here all Φ_n are clearly nonexponential and already for $q = 0.01$ a description solely in terms of $\Phi_{1,n}$ turns out not to be satisfactory; one has to use at least $\Phi_{2,n}$ in order to get a good picture of what happens during the first three decades of the decay. More complex even is the situation for $q = 0.05$: there $\Phi_{1,n}$ can be employed only over the first decade (as a rough approximation); also $\Phi_{2,n}$ captures only part of the picture, given that it does not approximate anymore Φ_n strictly from above. While the region where this happens may not be of experimental relevance (and still under the proviso that our numerical procedure may have its limitations) we can, on the basis of our preliminary results, state that for large n the behavior of the cumulants is by far more complex on SWNs than on regular lattices or on Cayley trees. In fact, on inhomogeneous lattices complexity may be very high.[58,59] Also the $\Phi_{3,n}$ and $\Phi_{4,n}$ approximants are not of much help, since they start their (unphysical, divergent) behavior before three decades of the decay are reached. Similar situations[8] were also encountered in one-dimension; there, however, one may help oneself through the knowledge[1] of a closed form for Φ; for SWNs, additional analyses are clearly necessary.

4. Conclusions

In this work we have focused on energy transfer and trapping on small world networks (SWNs). As discussed in the work, such SWNs are new classes of random graphs, which allow to combine in a judicious manner ordered and disordered aspects of realistic materials. As stressed above, such SWNs are akin in spirit to situations encountered in polymeric media.

Trapping on SWNs displays a series of features which interpolate between pure order and pure disorder. For a small density p of additional links (AL) the decay follows for some time the structure imposed by the parent lattice. Nonetheless, one observes in the decay behavior at longer times a crossover to a situation typical for an open tree;[46] this is due to the fact that the excitation uses increasingly steps along the ALs. A third decay domain would open up at very long times, where again the underlying geometry gets to be important;[46] we did not discuss this domain here, because we wanted to focus on the experimentally accessible decay regions.

The crossover behavior found here for trapping on SWNs is quite reminiscent of the transition in trapping behavior on regular lattices when going from low (small d) to high (large d) dimensions. The crossover is also akin to what happens in ultrametric spaces (UMS), when increasing the temperature T; namely, at higher T the decay law gets to be more regular and can be more readily described through S_n, the mean number of distinct sites visited by the walker in n-steps. In lower dimensions and at lower temperatures this is no more the case; the full distribution of distinct sites visited (and not only its mean value) matters. Starting here from a ring, on which we added ALs, several aspects of the decay are of importance: For large p, as mentioned, the decay gets to be very regular so that (at least for a very low density of traps) it becomes close to exponential; furthermore even in the region where such a decay is nonexponential, its form may still be well approximated through $\Phi_{1,n}$, i.e. with help of S_n. On the other hand, at very low AL densities, the 1D behavior persists for a long time. Of particular interest is the intermediate region of medium AL density, where the decay forms are quite complex: Their approximation through cumulant forms gets to be inaccurate at a rather early stage; our preliminary calculations show that this is due to the higher cumulants, which (distinct from the previous findings for regular lattices) do not necessarily stay positive. We thus view trapping on SWNs as a rather complex problem, which certainly deserves further study.

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