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Distribution Functions for Random Walk Processes on Networks: An Analytic Method

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A novel analytic method for deriving and analyzing probability distribution functions of variables arising in random walk problems is presented. Applications of the method to quasi-one-dimensional systems show that the generating functions of interest possess simple poles, and no branch cuts outside the unit complex disk. This fact makes it possible to derive closed formulas for the full probability distribution functions and to analyze their properties. We find that transverse structures attached to a one-dimensional backbone can be responsible for the appearance of power laws in observables such as the distribution of first arrival times or the total current moving through a (model) photoexcited dirty semiconductor (our results compare well with experiment). We conclude that in some cases a geometrical effect, e.g., that of a transverse structure, may be indistinguishable from a dynamical effect (long waiting time); we also find universal shapes of distribution functions (humped structures) which are not characterized by power laws. The role of bias in determining properties of quasi-one-dimensional structures is examined. A master equation for generating functions is derived and applied to the computation of currents. Our method is also applied to a fractal structure, yielding nontrivial power laws. In all finite networks considered, all probability distributions decay exponentially for asymptotically long times.

KEY WORDS: Random walk; probability distribution functions; first passage time; scaling.

1. INTRODUCTION

In recent years, with the realization that many disordered systems have interesting geometric structures, some of which have been coined fractals, it has become clear that random walks[1,2] on such systems provide a

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[2] For a relatively recent review with some historical background see Ref. 2.

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worthwhile research topic. These systems do not enjoy the simplicity of periodic lattice structures; however, they exhibit many exotic properties and new physical phenomena.[3] The extensive literature in such fields as percolation[4] or flow through porous media[5,6] is merely a part of the work in this domain.

Since nontrivial geometry plays an important role in the field of disordered systems, it is of crucial importance to differentiate between purely geometric effects and dynamical ones, whenever possible. For example, the difference between diffusion in a periodic system and in an irregular one is a geometric effect,[7,8] since the microscopic mechanisms are (in many cases) the same in both systems. As shown below, one may obtain geometrical effects that mimic dynamical effects. In such cases the distinction might be difficult to make.

Investigations of properties of random walks have mainly concentrated on mean quantities, such as mean first passage times and low moments of various observables. Detailed distribution functions (such as the distribution of first arrival times) have rarely been presented in analytical form. Obviously, full distribution functins are measurable in many cases and are of both physical and mathematical interest.

In the present work, we present methods for deriving full distribution functions of various physical quantities. Our formulas are analytic and we show how one can use them to deduce asymptotic properties such as various power laws characterizing correlations and spectra. The method and terminology used below rely heavily upon Ref. 9, in which a method for deriving generating functions was presented. Most of the applications below are to quasi-one-dimensional systems which are not lattice systems. Applications to higher dimensional systems will be presented in future publications.

The structure of this paper is as follows. In Section 2 we present the framework of the method for calculating distribution functions. In Section 3 we calculate the distribution functions for first passage time and first return time on a segment. Section 4 generalizes the results obtained for the simple segment to the case of a segment with dangling bonds. In Section 5 we evaluate the role of bias on the diffusion process and compare some of our results to an experiment. In Section 6 we present a brief summary of the results and relate to future work. Since some of the computations are rather lengthy, we present them in Appendices. Appendices A and B contain the detailed calculations leading to the results of Section 3 and 4. Appendix C contains computational details related to Section 5.

2. DEFINITIONS AND ASPECTS OF THE METHOD

In this section we review the main features of the method described in Ref. 9. As there, we restrict ourselves to nearest neighbor hopping in discrete time. We consider networks composed of connected segments (each of which is composed of a finite number of points; cf. Fig. 1).

The following quantities[9] are called elementary probability distributions:

1. $X_r(n)$: the probability to stay for n (time) steps at a point where the staying probability per step is r. Obviously: $X_r(n) = r^n$.
2. $T_{AB}(n)$: the probability to leave a point A on the first step and reach point B, for the first time, in n steps without ever returning to A.
3. $Q_{AB}(n)$: the probability to leave a point A on the first step and return to A for the first time without ever reaching B.

The probability to leave a point A and reach point B, for the first time, in n steps (the walker is allowed to return to A as often as desired) is denoted by $G_{AB}(n)$ and is not "elementary": it is expressible in terms of the elementary probability distributions.[9]

The generating function associated with a probability distribution $P(n)$ is

$$F(z) \equiv \sum_{n=0}^{\infty} z^n P(n) \tag{2.1}$$

The sum of probabilities $P(n)$ does not exceed unity, so that (2.1) is an absolutely convergent series on the unit disk. In principle, $P(n)$ can be derived from $F(z)$ by the use of Cauchy's theorem. For the sake of

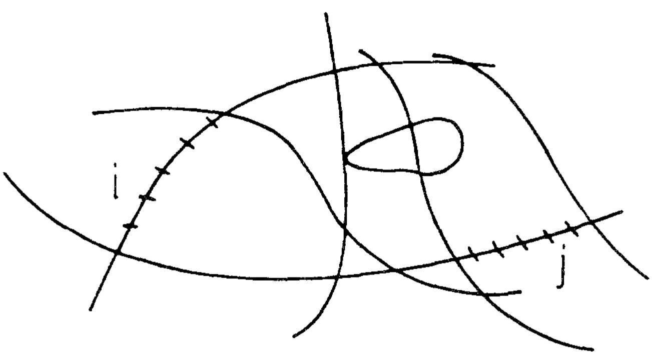

Fig. 1. A "random" network of points. The lines are drawn to guide the eye.

definiteness (and without limiting the generality of our results) we have chosen to assume that the probability of hopping from a point to any of its nearest neighbors in a single step is 1/4; it is zero for points other than itself and its nearest neighbors (we thus allow at most four nearest neighbors). When biased walks are considered, this assumption is modified.

It is easy to see that the generating function corresponding to staying at a point that has k nearest neighbors[9] is

$$X_{1-k/4}(z) = \frac{1}{1 - z(1 - k/4)}$$

Other generating functions are presented in Ref. 9. When no confusion can arise, we shall denote the probability function and its corresponding generating function by the same letter.

3. DEMONSTRATION IN A SIMPLE CASE: RANDOM WALK ON A SEGMENT

The problem is to calculate the probability distribution $P_N(n)$ for a walk starting at one end (point "0") of a segment of length N (i.e., having $N+1$ points) and reaching its other end (point "N") in n steps under given restrictions (see Fig. 2). We define the following functions: $t_N = T_{0,N}$ and $q_N = Q_{0,N}$. We found in Ref. 9 that

$$q_{N+1} = \left(\frac{z}{4}\right)^2 \frac{X_{1/2}}{1 - X_{1/2} q_N} \tag{3.1}$$

and

$$t_N = \left[\frac{(q_{N+1} - q_N)(1 - X_{1/2} q_N)}{X_{1/2}}\right]^{1/2} \tag{3.2}$$

Hence

$$t_N = \frac{z}{4}\left(1 - \frac{q_N}{q_{N+1}}\right)^{1/2} \tag{3.3}$$

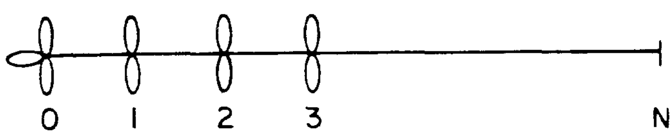

Fig. 2. The straight segment. Each loop attached on the sites indicates the (1/4) probability to stay.

The solution[9] of Eq. (3.1) is

$$q_N = \left(\frac{z}{4}\right)^2 X_{1/2} \frac{\lambda_+^{N-1} - \lambda_-^{N-1}}{\lambda_+^N - \lambda_-^N}$$

where

$$\lambda_\pm = \frac{1 + (1-4a)^{1/2}}{2} \tag{3.4}$$

$$\Lambda = \frac{\lambda_-}{\lambda_+} \tag{3.5}$$

$$a = \left(\frac{z}{4} X_{1/2}\right)^2 \tag{3.6}$$

Note that Λ is a known function of the complex variable z. In terms of Λ

$$q_N = \frac{1}{X_{1/2}} \frac{\Lambda}{\Lambda + 1} \frac{1 - \Lambda^{N-1}}{1 - \Lambda^N} \tag{3.7}$$

It follows from Eq. (3.3) that

$$t_N = \frac{z}{4} \Lambda^{(N-1)/2} \frac{1-\Lambda}{1-\Lambda^N} \tag{3.8}$$

We now wish to analyze the structure of q_N and t_N in the z plane. First of all, we show that q_N and t_N possess no branch points in the complex plane. Clearly, $(X_{1/2})^{-1}$ is analytic in z. There are three possible "sources" of branch points in Eq. (3.8). Either Λ diverges or it has a branch point itself or $\Lambda = 0$ in the complex z plane (and $N-1$ is odd). The quantity t_N can have a branch point only if Λ has one. However, $\Lambda = 0$ implies, using Eq. (3.5), that $\lambda_- = 0$ or $\lambda_+ = \infty$. Using Eq. (3.4), we find that $a = 0$ or $a = \infty$ is implied, respectively. Equation (3.6) implies $z = 0$ and $z = 2$, respectively. However, $z = 0$ cannot be a branch point because $q_N(z)$ and $t_N(z)$ are analytic inside the unit disk. When $z \to 2$, $a \to \infty$, and $\Lambda \to -1$ [from Eqs. (3.4) and (3.6)], contrary to the assumption $\Lambda = 0$. Hence $\Lambda \neq 0$ in the complex z plane. It is also impossible for Λ to diverge, since Eqs. (3.4) and (3.5) then imply that $\lambda_+ = 0$, which can happen only for $z = 0$, where we know that q_N and t_N are analytic. Finally, we have to check whether Λ itself has a branch point. Upon inspecting Eqs. (3.4)–(3.6), we see that Λ can have a branch point only when $a = 1/4$. It follows from Eq. (3.6) that at this point $z = 1$. The function (a) is analytic at $z=1$. In the neighborhood of this point we have $z=1+\varepsilon e^{i\phi}$, where $\varepsilon \ll 1$, and

$$\lambda_{\pm}=1 \pm [-4a'(1)\,\varepsilon e^{i\phi}]^{1/2} \tag{3.9}$$

Upon a rotation around $z=1$ from $\phi=0$ to $\phi=2\pi$, we have $\Lambda \to \Lambda^{-1}$. Since q_N and t_N are invariant under this transformation, they have no branch points. The only possible singularities of $q_N(z)$ and $t_N(z)$ in the complex plane are poles. When $z \to \infty$, $\Lambda \to 1$ nd hence q_N and t_N are bounded for large enough z. This means that contour integration of q_N/z^N and t_N/z^N for contours with z large enough tend to zero as $z \to \infty$ if $n>1$.

The poles of t_N are located on the unit circle in the Λ *plane*:

$$\Lambda_p = e^{2i\pi p/N}, \qquad p=1,\ldots, N-1 \tag{3.10}$$

and Λ itself has no pole. When $z \to \infty$, then $\Lambda \to 1$ and $t_N \to z/4N$. Consequently, application of Cauchy's theorem, when $F(z)$ is replaced by $t_N(z)$ or $q_N(z)$, can be performed by considering the poles *outside* the unit disk. When $N>1$ the "contour at infinity" makes no contribution. The technical details are presented in Appendix A. The result is

$$t_N(n)=\frac{1}{2N}\sum_{p=1}^{N-1}(-)^{p+1}\cos^{2n}\left(\frac{\pi p}{2N}\right) tg^2\left(\frac{\pi p}{2N}\right) \tag{3.11}$$

For large n, $t_N(n)$ is dominated by the first term in the sum, i.e., by the pole closest to unity. Approximating the cosine by an exponential (for $N \gg 1$), we obtain for large n

$$t_N(n)=\frac{1}{2N}\left(\frac{\pi}{2N}\right)^2 e^{-(\pi/2N)^2 n} \tag{3.12}$$

which is a valid approximation for $n \gg N^2$.

Figure 3 presents a plot of t_{100} versus n and the asymptotic exponential decay is indeed observed [the dotted points correspond to Eq. (3.12)].

Next, we wish to derive the probability distribution $G_N(n)$ for the first passage time from "0" to "N." As shown in Ref. 9,

$$G_N=\frac{X_{3/4}t_N}{1-X_{3/4}q_N} \tag{3.13}$$

Substituting the expressions for t_N and q_N [Eqs. (3.7) and (3.8)] into Eq. (3.13), we have for G_N in the variable $\mu \equiv \sqrt{\Lambda}$

$$G_N=\frac{z}{4}\frac{\mu^{N-1}(1+\mu)^3}{1+\mu^{2N+1}} \tag{3.14}$$

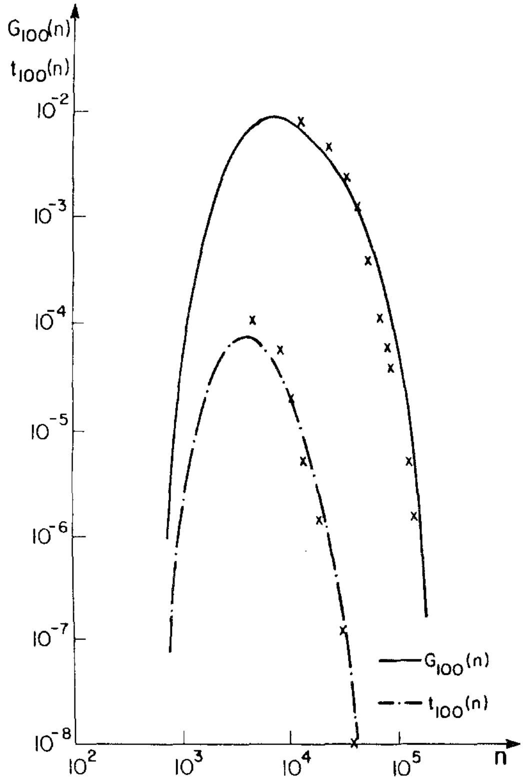

Fig. 3. Plot of G_{100} and t_{100} versus n for the straight segment (thick lines). The crosses are the asymptotic values as given by Eqs. (3.15) and (3.20). The asymptotic approximations are seen to agree reasonably well with the exact results for $n \geqslant \langle n \rangle$ [i.e., $\langle n \rangle \approx 2/3N^2$ for G_N and $\langle n \rangle \approx 2N(N+1)$ for t_N].

The poles of G_N are located at

$$\mu = -\exp\frac{2\pi i p}{2N+1}, \qquad p = 1,\ldots, 2N \tag{3.15}$$

As before, G_N has no other singularities but poles in the complex z plane. In this case we find (see Appendix A)

$$G_N(n) = \frac{(-)^N}{2N+1} \sum_{p=1}^{2N} (-)^p \sin^{2n} \frac{\pi p}{2N+1} \cos \frac{\pi p}{2N+1} \tag{3.16}$$

The leading term for $n \gg N$ comes from the pole corresponding to $p = N$ and $p = N+1$ (which is the same pole). Asymptotically

$$G_N(n) = \frac{\pi}{4N^2} e^{-(\pi/4N)^2 n} \tag{3.17}$$

(see the dotted points in Fig. 3). The slower exponential decay (with respect to that of t_N) is due to the fact that the walker is allowed to return to the origin. The mean first passage time (MFT) is $\langle n \rangle = 2N(N+1)$. This result is obtained by using Eq. (3.16) or directly (see Ref. 9).

4. RANDOM WALK ON A SEGMENT WITH DANGLING LOOPS

The system we consider is shown in Fig. 4. The "length" of each loop is m and the length of the segment is N. Define r_m to be the generating function corresponding to the probability to leave a site on the "backbone" on the first step, enter the loop of length m, and return to the same site for the first time at step "n." Since the walker may either return through the direction of entry into the loop or go all the way around the loop, we obtain

$$r_m = 2(q_m + t_m) \tag{4.1}$$

Next, define $Y_m(z)$ to be the generating function corresponding to the probability to go from a site on the backbone back to itself, without leaving to another site on the backbone. Summing over all multiple excursions into the loop, we have

$$Y_m = 1 + r_m + (r_m)^2 + \cdots = 1/(1 - r_m) \tag{4.2}$$

Note that for $m = 1$, we have $r_m = 2t_1 = z/2$ and $Y_1 = X_{1/2}$, which reduces to the previous case (i.e., no dangling loop).

The recursion relations for Q_N and T_N are the same as Eq. (3.1) and (3.2) with $X_{1/2}$ replaced by Y_m (at the first step the walker must go to site "1"). The recursion relations now read

$$Q_N = \left(\frac{z}{4}\right)^2 \frac{Y_m}{1 - Y_m Q_N} \tag{4.3}$$

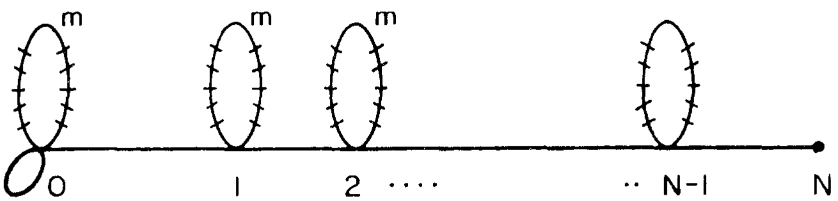

Fig. 4. The straight segment with dangling bonds. The extra little loop at the origin (site "0") represent the staying probability there (1/4 in the absence of bias).

and

$$T_N = \left[\frac{(Q_{(N+1)} - Q_N)(1 - Y_m Q_N)}{Y_m} \right]^{1/2} \tag{4.4}$$

Define

$$a \equiv \left(\frac{z}{4} Y_m \right)^2 \tag{4.5}$$

Then define $\lambda_\pm$ and Λ as in Eqs. (3.6) and (3.7). Q_N and T_N are given by expressions identical to Eqs. (3.7) and (3.8). Consequently,

$$\Lambda = \frac{1 - (1 - 4a)^{1/2}}{1 + (1 - 4a)^{1/2}} \tag{4.6}$$

It follows from Eqs. (4.5) and (4.6) that

$$\frac{z}{4} Y_m = \frac{\sqrt{\Lambda}}{\Lambda + 1} \tag{4.7}$$

As before, the only singularities of T_N are on the unit circle in the Λ plane. The location of the pth pole in the z plane follows from the expression for Y_m in (A.10) and Eqs. (3.10). Using Eq. (4.7), we obtain

$$2 \sin^2(\pi p/2N) = \sin \theta \operatorname{tg}(m\theta/2) \tag{4.8}$$

where θ is defined by

$$z = \frac{1}{\cos^2(\theta/2)}$$

Equation (4.8) has m real solutions (or $m-2$ solutions, depending on the lhs). This fact is exhibited in Fig. 5, which shows the rhs of Eq. (4.8) and its intersections with a fixed value of the lhs of this equation. It can be proven (see Appendix A) that Eq. (4.8) has only one complex solution at $\theta = \pi + iy$ and $y = O(1/m)$ for large enough m. Since $\theta = \pi$ corresponds to $z = \infty$, this solution is not relevant for large m and n (see Appendix B). Since p can assume $N-1$ different values, the number of solutions of Eq. (4.8) equals $(N-1)(m+1)$. It is important to note that $z = 1$ is not a pole. Using the pole structure, we can now evaluate the distribution function. The result for $T_N(n)$ is [see Appendix A for a derivation of expressions for $Q_N(n)$ and $G_N(n)$]:

$$T_N(n) = \frac{1}{4N} \sum_{p=1}^{N-1} \sum_{\{\theta_p\}} (-)^{p+1} \sin^2 \frac{\pi p}{N} \cos^{2(n-1)} \frac{\theta_p}{2} \operatorname{tg} \frac{\theta_p}{2} \times \frac{1 + \cos m\theta_p}{m \sin \theta_p + \sin m\theta_p \cos \theta_p} \tag{4.9}$$

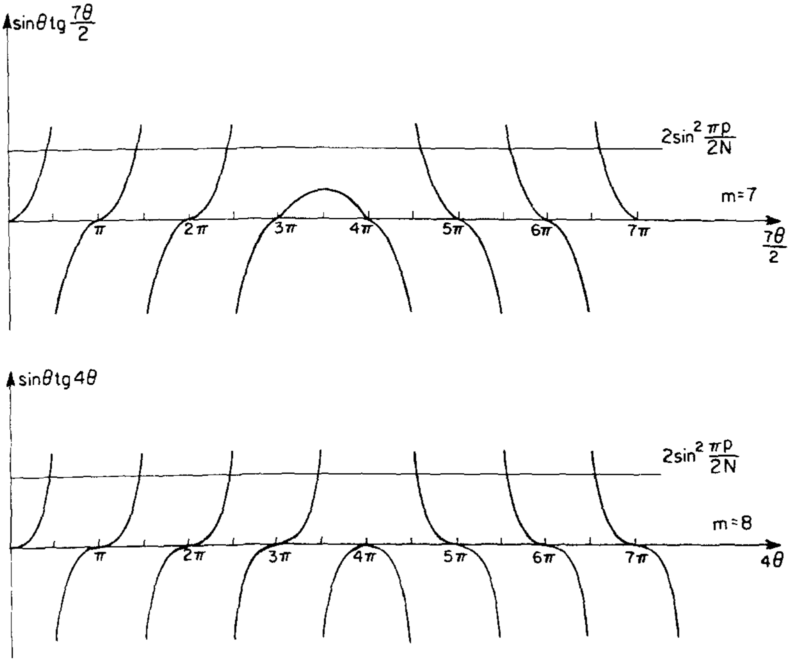

Fig. 5. Plot of the "pole equation" [Eq. (4.8)] for $m = 7, 8$. The horizontal line equals $2\sin^2(\pi p/2N)$.

where $\{\theta_p\}$ is the set of solutions of Eq. (4.8). Substituting $m = 1$ (no loops), we recover Eq. (3.11).

Obviously Y_m is the generating function for the mean waiting time distribution $\Psi_m(n)$ [denoted $\Psi(t)$ by Montroll and Weiss[14]]. The mean first passage time $\langle n \rangle$ follows in accordance with the result in Ref. 9, and the mean waiting time $\bar{t}$ is

$$\bar{t} = 2m - 1 \tag{4.10}$$

Note the linear dependence of the waiting time on the length of the loop m.

Figure 6 contains plots of G_N as a function of n for several large values of m [see Eq. (A.18)]. $G_N(n)$ has been normalized by its value at the MFT. Note that hump structure of G_N, which shows up distinctly for $m = 100$ ($N = 10$). It exists for higher values of m, too. Asymptotically for $n \to \infty$ the decay of G_N is exponential, as expected. When one plots $G_N(n)$ for several values of m keeping the parameter $\alpha = m/N^2 = \bar{t}/\langle n \rangle$ fixed, the resulting shapes are similar in the region of $\langle n \rangle$ (see Fig. 7). When $\alpha \ll 1$ the shape of G_N is similar to the one obtained in the absence of dangling bonds. The

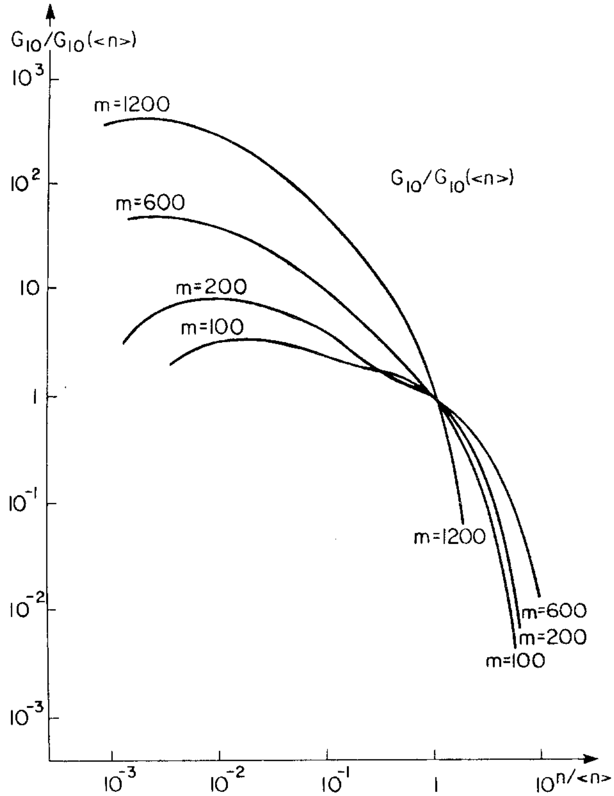

Fig. 6. Plot of G_{10} for $m = 100, 200, 600, 1200$ normalized by their MFT value versus $n/\langle n \rangle$.

humped structure appears for $\alpha = 1$. When $\alpha \gg 1$ this structure disappears. It is interesting to note that at $n = \langle n \rangle$ we have a transition in the slope, from a plateau-like to an exponentially decreasing behavior. In Appendix B we present an asymptotic analysis of $T_N(n)$ and $G_N(n)$. In the $n \to \infty$ asymptotic regime, these functions are dominated by the pole closest to unity, yielding a simple exponential decay. When $\alpha \gg 1$ we find an $n^{-3/2}$ decay (see Appendix B) even for intermediate values of n.

Next, consider $\Psi_m(n)$, the waiting time distribution. In Appendix A we give an expression for $\Psi_m(n)$ [see Eq. (A.20)]. Asymptotically in n (see Appendix B) we find

$$\Psi_m(n) \propto n^{-3/2} \qquad \text{for} \quad 1 \ll n \ll m^2 \tag{4.11}$$

Figure 8 is a plot of $\Psi_m(n)$. The asymptotic power-law behavior sets in already at very low n. Starting at $n = m^2$, the decay of $\Psi_m(n)$ is exponential.

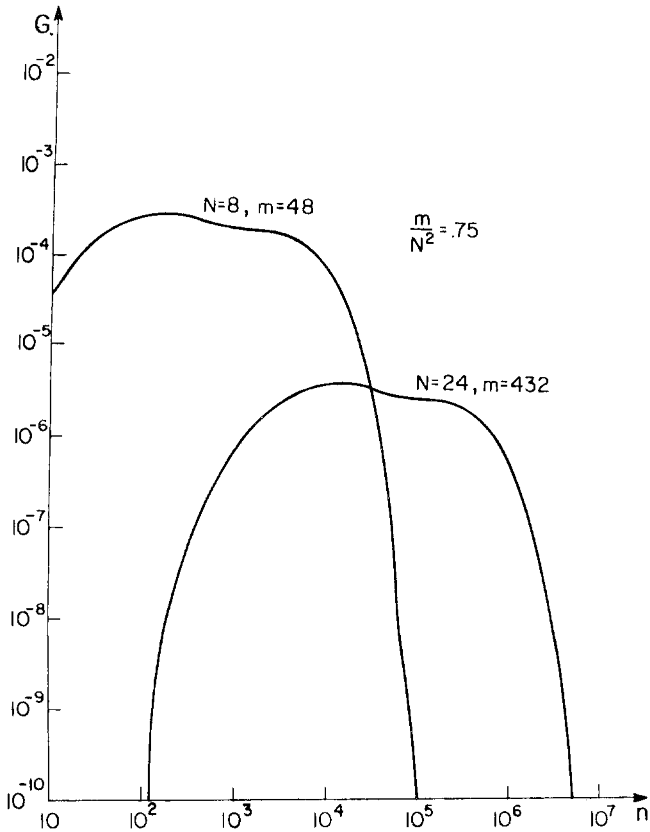

Fig. 7. Plots of G for $m/N^2 = 0.75$ for $N = 8$, $m = 48$ and $N = 24$, $m = 432$. Both curves have almost the same shape. Compare with the plateau region with the asymptotic values defining *is*, as derived in Appendix B.

A similar behavior for $\Psi_m(n)$ was previously obtained by Scher and Lax[15] in a different context in their work on transport in disordered solids. They obtain a waiting time distribution like Eq. (4.11) based on dynamical considerations, i.e., the spectrum of transition rates, whereas our result is of pure geometric nature. One sees that the same waiting time distribution can follow from a dynamical model and from a geometric construction. Thus, it might be difficult to disentangle geometric effects from dynamical ones. On the other hand, *if* one knows that very large dangling bonds are nonexistent in a given physical system, then a very dispersive waiting time distribution must be of a dynamical nature. In percolation clusters this dispersion can be of geometrical origin.

It is interesting to note that the formulas for T_N and G_N [Eqs. (4.9) and (A.18)] (as functions of the variable θ) are independent (in form) of

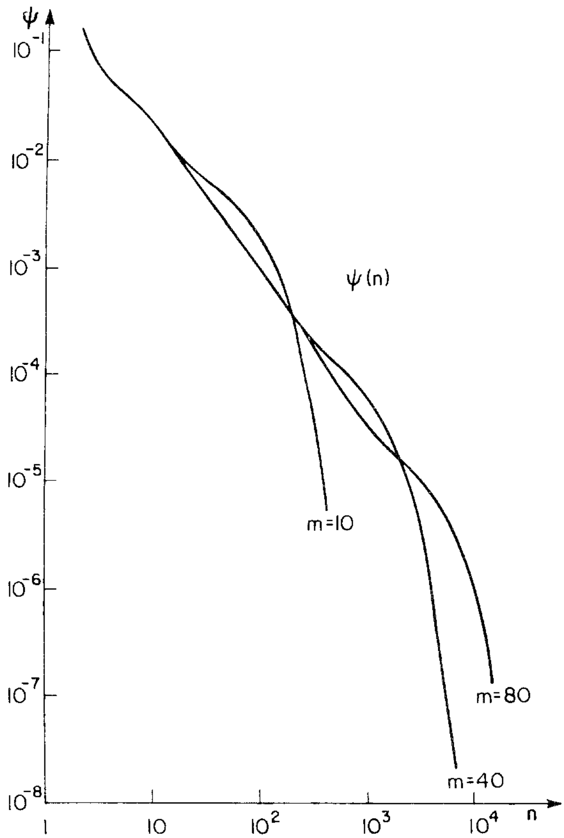

Fig. 8. Plots of $\Psi_m(n)$ for $m = 10$, 40, 80 versus n. The decay is clearly $n^{-3/2}$ with an exponential cutoff at $n = m^2$.

the transverse structure. The latter enters through the values of θ_p only, which for T_N are given by Eq. (4.8). The corresponding equation for G_N is given in Appendix A. Thus, our method enables us to compute these distribution functions for networks that have transverse structures different from the one presented here.

5. THE ROLE OF BIAS AND COMPARISON WITH EXPERIMENT

An example of an application of the method presented above is to the anomalous dispersion in amorphous solids.[16] An early experiment by Pfister[17] is now briefly described. A thin sample of amorphous solid (e.g., As_2Se_3) is placed between two electrodes. A light flash excites electron–hole pairs close to the anode. The resulting electron is absorbed by the anode and the hole moves toward the cathode. It is assumed that its motion is governed by hopping. Once the hole reaches the cathode, it is neutralized. The quantity measured in this experiment is $I(t)$, the *instantaneous total* current of the remaining holes in the sample (and not the number of holes arriving at the cathode per unit time).

Scher and Montroll[16] related the two slopes of the plot of $I(t)$ as a function of the time to a parameter α characterizing the tail of the waiting time distribution, i.e., $\Psi(t) \propto t^{-(1+\alpha)}$, where α is material-dependent (e.g., $\alpha = 0.8$ for TNF-PVK).[18] Let us assume that the waiting time distribution is of geometric (and not dynamical) origin and that it is due to the existence of the dangling bonds. Obviously, this assumption is not necessarily true. We present it as an extreme case in which geometry alone is responsible for the waiting time. In such a case α is geometry-dependent. As an illustration, we find [by computing $\Psi_m(n)$] that $\alpha = 1/4$ for the structure depicted in Fig. 9. This is a dangling bond with a loop-within-loop structure (all loops are of length m), which is self-similar (see Appendix A).

Consider a one-dimensional system (see Fig. 3) in which the hopping probabilities are p_1 in the direction of an external bias (to the right) and p_2 against the bias ($p_1 > p_2$). For definiteness we choose $p_1 + p_2 = 1/2$. Define b, the bias parameter:

$$p_1 = \kappa e^b \qquad \text{and} \qquad p_2 = \kappa e^{-b}; \qquad \text{hence} \quad \kappa = 1/(4 \cosh b)$$

Inside the dangling bond we assume for simplicity that the walk is not biased, i.e., the dangling bond is orthogonal to the direction of the field. In Pfister's experiment, typical numbers are: voltage across the system

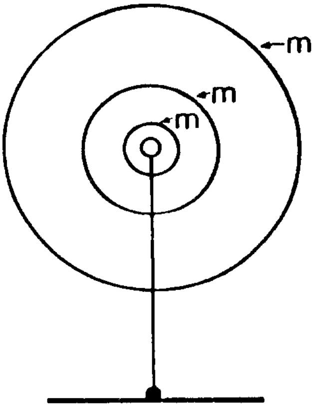

Fig. 9. The dangling "blob." The circles are of size m. Each site of the network is assumed to be attached to two such blobs.

$V = 1000$ V, thickness of the specimen $L \approx 91\ \mu$m, temperature $T \approx 300$ K. Therefore (assuming detailed balance), the bias parameter is $b = e\Delta V/\varepsilon kT$, where $\Delta V = V/N$ is the intersite potential difference, k is Boltzmann's constant, and ε is the dielectric constant. The intersite separation is[16] 1.17×10^{-6} cm, so that $N = 8 \times 10^3$ and $b \approx 1$. As shown in Appendix C, the bias is to be considered strong whenever $bN \gg 1$, which is obviously the case here. We now proceed to compute the total current corresponding to the above model.

The total instantaneous current is composed of contributions due to carriers that are on the backbone and hop to a neighboring site on the backbone. Hopping to the right (left) contributes a positive (negative) unit of current. Define $\bar{P}_k(t)$ to be the probability to be at time t at site "k" on the backbone. The total current $I(t)$ is obviously equal to $(p_1 - p_2)$ times the total number of carriers residing on the backbone at time t. Therefore $I(t)$ is given by

$$I(t) = N_c(p_1 - p_2)(\bar{P}_0 + \bar{P}_1 + \cdots + \bar{P}_{N-1}) + N_c \bar{P}_0 p_2 \tag{5.1}$$

where N_c is the number of carriers at $t = 0$. The second term on the rhs in Eq. (5.1) accounts for the fact that the carrier must leave the origin only to the right. The corresponding generating function $\bar{P}_k(z)$ satisfies the following "master equations" (A_m is defined in Appendix C):

$$\begin{aligned} \bar{P}_0 &= A_m + \bar{P}_1 p_2 z A_m \\ \bar{P}_k &= \bar{P}_{k-1} p_1 z Y_m + \bar{P}_{k+1} p_2 z Y_m, \qquad k = 1, \ldots, N-2 \\ \bar{P}_{N-1} &= \bar{P}_{N-2} p_1 z Y_m \end{aligned} \tag{5.2}$$

The rationale for these recursion relations is as follows. The probability to be at a point $0 < k < N$ is composed of the contributions of walkers who must have first reached its nearest neighbors on the backbone, then moved to "k" and (possibly) wandered through the loop attached to "k" and came back to it. At $k = N - 1$ there is no contribution of walkers arriving from $k = N$, because of the assumed absorbing boundary condition there. Since all walkers start at $k = 0$, by assumption, the probability $\bar{P}_0$ is composed of a part due to walkers who never left to $k = 1$ and those who did leave but eventually returned from $k = 1$ to $k = 0$. Define

$$\bar{P}_T = \bar{P}_0 + \bar{P}_1 + \cdots + \bar{P}_{N-1} \tag{5.3}$$

Summing Eqs. (5.2) and using the fact that $p_1 + p_2 = 1/2$, we obtain

$$\bar{P}_T = \frac{A_m + \bar{P}_1 p_2 z A_m - \bar{P}_1 p_2 z Y_m - \bar{P}_0 p_2 z Y_m - \bar{P}_{N-1} p_1 z Y_m}{1 - z Y_m/2} \tag{5.4}$$

We note that $\bar{P}_{N-1} p_1 z = G_N$. Using (C.13) and the first equation in (5.2), we have

$$\bar{P}_T = Y_m \frac{1-G_N}{1-zY_m/2} \tag{5.5}$$

We are interested in the current for large times ($t > N$) in the limit of very long dangling bonds. This can be accomplished by use of a Tauberian argument (see Appendix A.3). First, in Appendix A.3 we show that the large-t limit corresponds to small θ. We also find that [see Eqs. (A.10)]

$$(zY_m/2)^{-1} = 1 - \sin\theta \operatorname{tg}(m\theta/2)$$

where

$$z = e^{i\phi} = \frac{1}{\cos^2(\theta/2)}$$

The latter yields, for small θ (large t), $\theta \approx 2(i\phi)^{1/2}$. In the long-time limit, $\sin\theta = \theta$, but for times much shorter than the mean waiting time it is not *necessarily true* that $\operatorname{tg}(m\theta/2) = m\theta/2$. If m is large enough, $\operatorname{tg}(m\theta/2) = O(1)$. Note that $\operatorname{tg}(m\theta/2)$ is bounded in the range considered. Equation (A.2) ($\theta \equiv x + iy$) reads

$$\operatorname{tg}\frac{m\theta}{2} = \frac{\sin(mx) + i\sinh(my)}{\cos(mx) + \cosh(my)}$$

with nonvanishing denominator; since $\theta = 2(i\phi)^{1/2}$ and ϕ is real, then $y = (2\phi)^{1/2}$. In the small-θ limit, we find, following some algebra and using Eqs. (C.5), (C.6), and (C.10),

$$\Lambda \approx e^{-2b}\left(1 - 2\frac{\theta}{\operatorname{th} b}\cosh b \operatorname{tg}\frac{m\theta}{2}\right) \tag{5.6}$$

Hence, for small b, by Eq. (C.16),

$$\zeta \approx i\left(b + \frac{\theta}{\operatorname{th} b}\cosh b \operatorname{tg}\frac{m\theta}{2}\right)$$

Using Eq. (C.17) and defining

$$\beta \equiv \frac{\cosh b}{\operatorname{th} b}\operatorname{tg}\frac{m\theta}{2}$$

we obtain

$$\bar{P}_T = \frac{-2(1-\theta^2/4)}{\theta \operatorname{tg}(m\theta/2)} \frac{e^{(b+\theta\beta)} - e^{-b} - 2e^{-N\theta\beta}\sinh(b+\theta\beta)}{e^{(b+\theta\beta)} - e^{-b}} \tag{5.7}$$

It remains to evaluate the term $\bar{P}_0 p_2$ in (5.1). $\bar{P}_0$ is the generating function to stay at site "0" without leaving to "N." Using Eq. (C.12), we have

$$\bar{P}_0 = \frac{A_m}{1 - A_m Q_N} \tag{5.8}$$

Since $bN \gg 1$, we have from Eqs. (C.7) and (5.6)

$$Q_N \approx Y_m^{-1} \frac{A}{A+1}$$

and using Eq. (C.10),

$$Q_N \approx z(p_1 p_2 A)^{1/2}$$

Finally, using Eq. (C.13) and (5.6), we obtain

$$\bar{P}_0 p_2 \approx \frac{1}{z} \frac{1}{(1/2p_2)(zY_m/2)^{-1} - 1 - (1 - \theta\beta)}$$

$$\bar{P}_0 p_2 \approx \frac{1 - (\theta/2)^2}{(1/2p_2)^2} \left[1 + \theta \left(\frac{1}{2p_2} \operatorname{tg} \frac{m\theta}{2} - \beta \right) \frac{1}{2p_2 - 2} \right]$$

which for large t behaves like $t^{-3/2}$ since $\theta \approx \sqrt{\phi}$ and the factor $(=K)$ multipling θ is approximately constant. Thus, $\bar{P}_0 p_2 \propto 1 + K\sqrt{\phi}$ and it follows that $\bar{P}_0 p_2$ goes like $t^{-3/2}$ for (intermediately) large t. We consider two regimes: $N\theta/b \gg 1$ and $N\theta/b \ll 1$ (and $\beta \approx i/b$). When $N\theta/b \gg 1$, then $\bar{P}_T \propto 1/\theta$, i.e., $\bar{P}_T \propto t^{-1/2}$ for $t \gg (N/b)^2$. When $N\theta/b \ll 1$, then $\bar{P}_T \propto N/b(1 - \frac{1}{2}N\theta k)$, i.e., $\bar{P}_T \propto t^{-3/2}$ for $t \gg (N/b)^2$. Since $\bar{P}_0 p_2$ behaves like $t^{-3/2}$, the behavior of the total current is dominated by $\bar{P}_T$ and $I(t) \approx N_c \bar{P} V_T$. Thus

$$I(t) \propto \begin{cases} t^{-1/2} & \text{for} \quad t \ll (N/b)^2 \\ t^{-3/2} & \text{for} \quad t \gg (N/b)^2 \end{cases}$$

The "transition time" between the two power law regimes is thus proportional to $(N/E)^2$. Experimentally, the transition in the slope (which is used to operationally define the transit time in Ref. 16) is found to happen at $t \approx (N/E)^{2.2}$. Thus, our results agree reasonably well with experiment. The correction to the power 2 (i.e., 2.2) must follow either from a dynamical effect, such as the waiting time distribution, or the oversimplification involved in the model for the dangling bond. For example, the latter can be of fractal nature. The time at which the experimental curve starts to decay exponentially is, according to our theory, related to the mean waiting time (trapping time). The transit time so defined is *not* the mean first passage time $\tau(b)$ for a specific carrier. In Appendix C it is shown that

$$\tau(b) \approx N(2m-1)\coth b = (2m-1)(N/b)$$

which is m-dependent. The factor $(2m-1)$ is the mean waiting time in a dangling bond. The effective mean waiting time is thus field-dependent. The bias effectively reduces the time spent in the "traps." It is interesting to note the role of the length of the dangling bond m in this problem. As long as typical times are much shorter than the time necessary to "sense" the length of the dangling bond (i.e., the typical time to move through its entire length), the physics cannot possibly depend on the length m. As a result, the dangling bond is effectively infinite in length and the dispersion has no specific time scale: it is a power law. When one considers larger times, the length of the dangling bond acts as a cutoff on the residence times in it and one crosses over to exponential decay of the transit time distribution. This geometric picture seems to be relevant to many other physical systems, such as porous media. However, as noted above, we cannot rule out a dynamical, nongeometric, origin of the waiting time distribution, especially in solid-state physics. Both approaches lead, however, to similar results, which means that in this case, as we mentioned before, it is very difficult to disentangle geometry from dynamics (without additional information).

6. SUMMARY AND DISCUSSION

We have presented a method to compute the full distribution functions for random walks on networks. The technical part involves an analysis of the analytic properties of the generating functions corresponding to these walks. The key observation in this analysis is that a contour integration (aimed at finding the distribution function) on the unit circle can be regarded as an integration on the boundary of the domain outside the unit disk. In this domain the generating functions normally have only simple poles (we found no cuts, but we encountered examples of multiple poles), which makes the computation of the distribution fuctions possible. The result in every case we have presented is a closed formula for the distribution function. The type of formulas we have obtained can be further analyzed to obtain simple expressions which are uniformly valid for all times; this analysis, however, is left to a future publication.[10] We have seen that for long enough times, all distribution functions decay exponentially. The rate of this decay depends on the location of the pole, of the corresponding generating function, which is closest to the origin. When a random walker on network can be delayed for long times—either because of the existence of long dangling bonds or side structures—or because of long trapping times (waiting times), one expects a power law decay of the distribution for intermediate times. This power law decay is eventually cut off by the (truly) asymptotic exponential decay. The physical reason for the existence of the power law region is as follows: as long as the relevant times are much shorter than the relevant time scale (say, average trapping time) the value of this time is irrelevant.

We have also briefly considered the effect of bias on the distribution function and the influence of the dangling bonds in this case. The delaying effect due to the dangling bonds leads, as in the unbiased case, to power law regions in the distribution functions. The latter can show up as power laws in other observables, such as the total current in Pfister's experiment.

An important feature of the results presented is that the effect of dangling bonds can simulate the waiting time distribution of traps. This makes the distinction between dynamical and geometric effects quite subtle. Another feature of importance is the shape of the distribution function for first arrival. A comparison of our results with numerical simulations[12] done on fractal networks or experimental data shows that these shapes are quite "universal." We believe that the reason for this shape similarity is the fact that a percolating cluster has basically a single backbone "channel" and the rest of the bonds act as dangling bonds. Thus, in spite of the fact that we have not computed random walks on fractal networks, the results are similar. A computation of random walks on fractal networks, as well as other applications, are in progress.

APPENDIX A. CALCULATION OF THE PROBABILITY DISTRIBUTIONS

In this Appendix we calculate the probability distributions $T_N(n)$, $G_N(n)$, and $Q_N(n)$ for a segment with a dangling bond of length m (Fig. 4). The case of no dangling bonds corresponds to $m=1$.

First, we need the following results:

For C_p such that $0<C_p<2$ the only *nonreal* solution of the equation [see Eq. (4.8), where $C_p=2\sin^2(\pi p/2N)$]

$$C_p=\sin\theta\,\mathrm{tg}(m\theta/2) \tag{A.1}$$

is $\theta=\pi+iy$, where y is the real solution of

$$C_p=\frac{\sin y\sinh(my)}{(-)^m+\cosh(my)}$$

The proof is as follows. Define $\theta=x+iy$. Hence

$$\mathrm{tg}\frac{m\theta}{2}=\frac{\sin(mx)+i\sinh(my)}{\cos(mx)+\cosh(my)} \tag{A.2}$$

Define

$$\bar{C}_p=C_p[\cos(mx)+\cosh(my)]$$

Therefore

$$\bar{C}_p = \sin x \sin(mx) \cosh y - \cos x \sinh y \sinh(my)$$
$$0 = \sin x \sinh(my) \cosh y + \cos x \sinh y \sin(mx) \tag{A.3}$$

The quantity $\bar{C}_p$ is always positive, since $C_p > 0$ by assumption and $\cosh(my) > 1$ for any real y. Assuming $\sin x \neq 0$, we obtain from (A.3)

$$\frac{-\sin(mx)}{\sin x} \cos x = \frac{\sinh(my)}{\sinh y} \cosh y > 0$$

because $[\sinh(my)/\sinh y] > 0$. Defining $F(x) = -\sin(mx) \operatorname{cotg} x$, we have $F(x) > m$. At an extremum x_0 of $F(x)$ we have

$$\sin(mx_0) = m \cos(mx_0) \cos x_0 \sin x_0$$

Hence [since $F(x) > m$ for any $x \neq 0 \pmod{\pi}$]

$$F(x_0) = -m \cos(mx_0) \cos^2 x_0 > m$$

which is impossible. We conclude that $\sin x = 0$. Thus $x = 0$ or $x = \pi$ ($0 \leqslant x < 2\pi$ by definition). But for $x = 0$ it follows from Eq. (A.3) that C_p is negative, contrary to our assumption. Thus, $x = \pi$ only is allowed.

In the following, Eq. (A.1) is sometimes called the "pole equation."

A1. Calculation of $T_N(n)$ and $Q_N(n)$ for a Segment with Loops

In this case the "pole equation" reads $C_p = 2\sin^2(\pi p/2N)$. Applying Cauchy's theorem to (3.8), we have

$$T_N(n) = \frac{-1}{2\pi i} \int_\Gamma \frac{1}{4z^n} \Lambda^{(N-1)/2} \frac{1-\Lambda}{1-\Lambda^N}\, dz \tag{A.4}$$

where Γ is a contour enclosing the origin. The minus sign is here because the domain is *outside* the unit disk. A contour at ∞ has vanishing contribution (except when $n = 1$), since Λ is bounded everywhere. The only singularities in the above integrand, which we denote by I, are poles (see text). The residue $\operatorname{Res} I$ at a pole in the z plane is given by

$$\operatorname{Res} I = \lim_{z \to z_{\text{pole}}} \left(\frac{z - z_{\text{pole}}}{\Lambda - \Lambda_{\text{pole}}} \right) \cdot I \cdot (\Lambda - \Lambda_{\text{pole}}) \tag{A.5}$$

where

$$\Lambda|_{\text{pole}} = e^{2\pi i p/N}, \qquad p = 1,..., N-1 \tag{A.6}$$

The rest of the calculation is lengthy but straightforward. It is convenient to define the variables μ, θ, and α: $z = 4\mu/(1+\mu)^2$, $\mu = e^{i\theta}$, and $\alpha = \frac{1}{4} z Y_m$.

Hence, from Eq. (4.7),

$$\alpha = \sqrt{\Lambda}/(\Lambda + 1) \tag{A.7}$$

Thus,

$$z = \frac{1}{\cos^2(\theta/2)} \tag{A.8}$$

By the chain rule of differentiation (subscripts denote derivatives)

$$z_A = z_\mu \mu_\alpha \alpha_A \tag{A.9}$$

Using Eqs. (4.2) and (A.8), we write Y_m as

$$Y_m = \frac{2/z}{1 - \sin\theta \,\mathrm{tg}(m\theta/2)} \tag{A.10}$$

Doing the chain derivative, we obtain the first factor in the rhs of Eq. (A.4). The factors in the rhs of Eq. (A.9) are given by

$$\begin{aligned} z_\mu &= 4\frac{1-\mu}{(1+\mu)^3} \\ \mu_\alpha &= \frac{1+\cos m\theta}{m\sin\theta + \cos\theta\sin m\theta}\,\frac{(1+A)^2}{2A}\, ie^{i\theta} \\ \alpha_A &= \frac{1}{2\sqrt{A}}\,\frac{1-A}{(1+A)^2} \end{aligned} \tag{A.11}$$

where we also used

$$\lim_{A \to A_{\text{pole}}} \frac{A_{\text{pole}} - A}{1 - A^N} = \frac{1}{N} e^{2\pi i p/N} \tag{A.12}$$

Summing over the contribution of all poles, we obtain Eq. (4.9). Similarly,

$$\begin{aligned} Q_N(n) = \frac{-1}{N} \sum_{p=1}^{N-1} \sum_{\{\theta_p\}} \sin^2\frac{\pi p}{N} \cos^{2(n-1)}\frac{\theta_p}{2}\,\mathrm{tg}\frac{\theta_p}{2} \\ \times \frac{1+\cos m\theta_p}{m\sin\theta_p + \sin m\theta_p \cos\theta_p} \end{aligned} \tag{A.13}$$

where $\{\theta_p\}$ is the set of solutions of Eq. (A.1) where $C_p = 2\sin^2(\pi p/2N)$.

A2. Calculation of $G_N(n)$ for the System with Loops

Let A_m be the generating function for leaving point "0" into the first loop and returning to "0" or staying at "0." A single incursion into the loop yields r_m [see Eq. (4.1)]. Summing over all the possible incursions, we have

$$\begin{aligned} A_m &= 1 + (r_m + z/4) + (r_m + z/4)^2 + \cdots \\ A_m &= \frac{1}{1-(r_m + z/4)} \end{aligned} \tag{A.14}$$

Note that A_m is the generalization of $X_{3/4}$ [see Eq. (3.13)]. The generating function for the first passage time is

$$G_N = \frac{A_m T_N}{1 - A_m Q_N} \tag{A.15}$$

Substituting $v \equiv \sqrt{A}$ [see Eq. (4.6)] and employing Eqs. (A.14), (3.7), and (3.8), we obtain

$$G_N = \frac{v^N(1 - v^2)}{1 + v^{2N+1}} \tag{A.16}$$

The poles are now located at

$$v_{\text{pole}} = -e^{2\pi i p/(2N+1)}, \qquad p = 1,..., 2N \tag{A.17}$$

We find

$$G_N(n) = \frac{(-)^N}{2N+1} \sum_{p=1}^{2N} \sum_{\{\theta_p\}} (-)^p \sin\frac{2\pi p}{2N+1} \sin\frac{\pi p}{2N+1} \cos^{2n}\frac{\theta_p}{2} \operatorname{tg}\frac{\theta_p}{2}$$
$$\times \frac{1 + \cos m\theta_p}{m \sin\theta_p + \sin m\theta_p \cos\theta_p} \tag{A.18}$$

where $\{\theta_p\}$ is the set of solutions of Eq. (A.1). From Eqs. (A.17) and (4.7)

$$C_p = 2\cos^2\frac{\pi p}{2N}$$

A3. Calculation of $\Psi_m(n)$

$\Psi_m(n)$ is the probability distribution corresponding to Y_m. We have

$$\Psi_m(n) = \frac{-1}{2\pi i} \int_\Gamma \frac{2}{z^{n+1}} \frac{1}{1 - \sin\theta \operatorname{tg}(m\theta/2)} dz \tag{A.19}$$

The calculation runs exactly as before, yielding

$$\Psi_m(n) = 2 \sum_{\{\theta\}} \cos^{2n+1}\frac{\theta}{2} \sin^2\frac{\theta}{2} \frac{1}{2\cos\theta + \frac{1}{2}m(1 + \sin^2\theta)} \tag{A.20}$$

$\{\theta\}$ is the set of poles of Y_m [see Eq. (A.10)]. We have

$$1 = \sin\theta \operatorname{tg}(m\theta/2)$$

Note that $\Psi_1(n) = (\frac{1}{2})^n$, as it should.

In Appendix B we show explicitly that $\Psi_m(n) \approx n^{-3/2}$ for large n. Since the generating function is the discrete Laplace transform (Poisson transform) of the corresponding waiting time distribution, we can use Tauberian arguments relating the small-ϕ behavior ($z = e^{i\phi}$) to the large-n behavior. Fof small θ, by Eq. (A.8) we have $\theta \approx 2(i\phi)^{1/2}$. If θ is small but $m\theta \gg 1$, then $\text{tg}(m\theta/2) \approx i$ [see definition of θ, Eq. (A.8)] and we obtain, using Eq. (A.11),

$$Y_m \approx 2(1 - \theta^2/4)(1 + i\theta) \approx 2[1 + 2i(i\phi)^{1/2}]$$

and consequently $\Psi_m(n) \approx n^{-3/2}$.

A4. A Self-Similar Dangling Bond

The purpose of this subsection is to show, in principle, how a transverse structure different from the loop can be introduced into the model. As an example of a self-similar bond that may have physical significance, consider the "iterative" (finite or infinite) blob of Fig. 9.

Denote by B_k the generating function of a single excursion into the kth iterated blob (k-circles): B_k is the generating function for a walk in which the walker enters the kth iterated blob and returns to the backbone for the first time. It is understood that the innermost blob is "terminated" by a fictitious bond (by virtue of our convention). Thus, B_1, corresponding to a "tadpole," is

$$B_1 = \left(\frac{z}{4}\right)^2 \frac{1}{1 - z/4 - r_m} \tag{A.21}$$

The recursion relation for B_k is obtained as follows. After the first step leaving the backbone the walker has the option to go back to the backbone, to enter the loop, or to proceed to the next iteration. Thus

$$\begin{aligned} B_{k+1} &= \left(\frac{z}{4}\right)^2 [1 + (r_m + B_k) + (r_m + B_k)^2 \cdots] \\ B_{k+1} &= \left(\frac{z}{4}\right)^2 \frac{1}{(1 - r_m) - B_k} \end{aligned} \tag{A.22}$$

Recall that $Y_m^{-1} = 1 - r_m$ [cf Eq. (4.2)].

Equations (A.21) and (A.22) completely define the system. Define

$$\gamma_\pm = \frac{1}{2}\left\{1 \pm \left[1 - \left(\frac{zY_m}{2}\right)^2\right]^{1/2}\right\} \tag{A.23}$$

The solution of the recursion relation (A.22) is

$$B_k = \left(\frac{z}{4}\right)^2 Y_m \frac{H_-\gamma_-^{k-1} + H_+\gamma_+^{k-1}}{H_-\gamma_-^k + H_+\gamma_+^k} \tag{A.24}$$

and the quantities H_+ and H_- are determined by B_1. Defining $x \equiv H_-/H_+$, we have

$$B_1 = \left(\frac{z}{4}\right)^2 Y_m \frac{x+1}{x\gamma_- + \gamma_+} \tag{A.25}$$

Comparing Eqs. (A.21) and (A.25), we find

$$x = \frac{1 - z/4 - \gamma_+}{\gamma \quad + z/4 - 1} \tag{A.26}$$

Note that in this case x is a function of z. Defining $\Gamma \equiv \gamma_-/\gamma_+$, we have

$$B_k = \left(\frac{z}{4}\right)^2 \frac{Y_m}{\gamma_+} \frac{1 + x\Gamma^{k-1}}{1 + x\Gamma^k} \tag{A.27}$$

The $\gamma_\pm$ in (A.23) has a branch point at $z=1$ because $Y_m(1)=2$. Under a rotation around $z=1$ (see Section 3) we have $\gamma_+ \leftrightarrow \gamma_-$, $\Gamma \to \Gamma^{-1}$, and $x \to x^{-1}$. Hence $B_k \to xB_k$. However, $x(1)=1$, which yields that B_k has no cut. The analog of Y_m for the kth iterated blob, denoted $Y_B(k, m)$, is

$$Y_B(k, m) = 1 + 2B_k + (2B_k)^2 + \cdots = 1/(1 - 2B_k) \tag{A.28}$$

(the factor 2 comes from the fact that two blobs are attached to each site). In the limit $k \to \infty$ we have from (A.28)

$$B \equiv \lim_{k \to \infty} B_k = (z/4)^2\, Y_m \gamma_\pm^{-1} \tag{A.29}$$

where $\Gamma < 1$ yields γ_+ and $\Gamma > 1$ yields γ_-.

The analog of Y_m for the (infinite) blob is denoted $Y_b(m)$:

$$Y_b(m) = \frac{1}{1 - (z/2)^2\, Y_m\{1 \pm [1 - (zY_m/2)^2]^{1/2}\}^{-1}} \tag{A.30}$$

$Y_b(m)$ is the probability of going from the backbone through the blob and back to the backbone (not necessarily for the first time). From (A.30) one can in principle derive the waiting time distribution by Cauchy's theorem. To find the poles, simply use $Y_b(m)$ in place of Y_m:

$$\frac{z}{4} Y_b(m) = \frac{\sqrt{\Lambda}}{\Lambda + 1} \tag{A.31}$$

The rest of the derivation is identical to the case of the simple dangling bond. In conclusion, we have shown on this example that any transverse structure whose generating function is known can be solved using our method. As we have done for the simple dangling bond, we use a Tauberian argument to find the large-n behavior ($n \ll m^2$, i.e., $\theta m \gg 1$) for the blob. First we calculate Γ for small θ. From Eq. (A.23), $\gamma_\pm \approx \frac{1}{2}[1 \pm (-2\theta i)^{1/2}]$. Since $\Gamma = \gamma_-/\gamma_+$, we find $\Gamma \approx 1 + 8e^{i\pi 7/8}\phi^{1/4}$ ($z = e^{i\phi}$) and $\Gamma < 1$ and the positive sign must be chosen in Eq. (A.30). Hence, using Eq. (A.31),

$$Y_b(m) \approx 2\{1 - 2[-i(i\phi)^{1/2}]^{1/2}\} \approx 2(1 + 2\phi^{1/4}e^{i\pi 7/8})$$

and the waiting time distribution for the blob behaves like $n^{-5/4}$ for (intermediately) large n.

APPENDIX B. ASYMPTOTIC ANALYSIS OF THE DISTRIBUTION FUNCTIONS

B1. Asymptotic Behavior of $G_N(n)$ and $T_N(n)$

For very large n the functions $G_N(n)$ and $T_N(n)$ decay exponentially since they are sums of asymptotically exponential functions (the pole closest to $z = 1$ dominates). In this Appendix we study the behavior of these functions in the moderately large-n region so that no single exponential dominates.

We propose to study the asymptotic behavior of T_N and G_N simultaneously. These quantities are given in Eqs. (A.18) and (4.9). We redefine the index of summation of G_N in order to bring its "pole equation" to a form similar to that of T_N. Define a new index of summation by performing the replacement $p \to p + N$. Then G_N reads [see Eq. (A.18)]

$$G_N(n) = \frac{1}{2N+1} \sum_{p=-N+1}^{N} \sum_{\{\theta\}} (-)^p \times \sin\left(\frac{\pi}{2+1/N} + \frac{\pi p}{2N+1}\right) \sin\left(\frac{\pi}{1+1/2N} + \frac{2\pi p}{2N+1}\right) \times \operatorname{tg}\frac{\theta}{2} \cos^{2n}\frac{\theta}{2} \frac{1+\cos m\theta}{2\cos\theta \sin m\theta + m \sin\theta}$$

For large N this is approximately equal to

$$G_N(n) \approx \frac{2}{2N+1} \sum_{p=1}^{N} \sum_{\{\theta\}} (-)^{p+1} \cos\frac{\pi p}{2N+1} \sin\frac{2\pi p}{2N+1} \times \operatorname{tg}\frac{\theta}{2} \cos^{2n}\frac{\theta}{2} \frac{1+\cos m\theta}{2\cos\theta \sin m\theta + m\sin\theta}$$

G_N and T_N are approximately equal to

$$P(n) \approx \sum_{p=1}^{N} \sum_{\{\theta\}} (-)^{p+1} F(p) \cos^{2n}\frac{\theta}{2} \operatorname{tg}\frac{\theta}{2} \frac{1+\cos m\theta}{m \sin\theta + \sin m\theta \cos\theta} \tag{B.1}$$

with a "pole equation" of the form

$$C_p = \sin\theta \operatorname{tg}(m\theta/2) \tag{B.2}$$

where for T_N [see Eq. (4.9)]

$$F(p) = \frac{1}{4N} \sin^2\frac{\pi p}{N} \tag{B.3}$$

and

$$C_p = 2\sin^2\frac{\pi p}{N} \tag{B.4}$$

whereas for G_N [see Eq. (A.18)]

$$F(p) = \frac{2}{2N+1} \sin\frac{2\pi p}{2N+1} \cos\frac{\pi p}{2N+1} \tag{B.5}$$

and

$$C_p = 2\sin^2\frac{\pi p}{2N+1} \tag{B.6}$$

which for large N reduces to Eq. (B.4). Clearly, for large n, only small values of θ contribute to the sum in Eq. (B.1). The "pole equation" (B.4) reads for small θ

$$C_p \approx \theta \operatorname{tg}(m\theta/2) \tag{B.7}$$

which, by defining

$$x = \tfrac{1}{2}m\theta \tag{B.8}$$

and

$$K_p = \tfrac{1}{2}mC_p \tag{B.9}$$

can be written as

$$K_p \approx x \operatorname{tg} x \tag{B.10}$$

From Fig. 5 it is seen that the solutions x_s take the form

$$x_s = \pi s + \alpha_s \pi/2 \tag{B.11}$$

where s is a nonnegative integer. We have $0 < \alpha_s < \pi/2$ and x_s is the solution corresponding to s. Note that if $s = O(m)$, then by Eq. (B.11), x_s is $O(m)$ and by Eq. (B.8), $\theta = O(1)$, which does not contribute to the sum in Eq. (B.1) because it contributes an exponentially small correction. Equation (B.10) is

$$K_p \approx (\pi s + \alpha_s \pi/2)\,\mathrm{tg}(\alpha_s \pi/2) \tag{B.12}$$

The factor to the right of $\cos^{2n}(\theta/2)$ in Eq. (B.1) is

$$\frac{\pi}{m}\left(s + \frac{\alpha_s}{2}\right) \frac{1 + \cos \alpha_s \pi}{\pi(2s + \alpha_s) + \sin(\alpha_s \pi/2)} \tag{B.13}$$

We consider two regimes of K_p: $K_p \gg 1$ and $K_p \ll 1$. The first case occurs if $mC_p \gg 1$ [see Eq. (B.9)]. If $p = O(N)$ [the index of summation in Eq. (B.1)], then by Eq. (B.4), $C_p = O(1)$ and $K_p \gg 1$. Thus, in the sum of Eq. (B.1) there are always terms with $K_p \gg 1$. If $m/N^2 \gg 1$ [see again Eqs. (B.1) and (B.4)–(B.9)], then $K_p \ll 1$ is impossible. Hence, if $m/N^2 \gg 1$, only terms with $K_p \gg 1$ contribute to the sum. If $m/N^2 = O(1)$ or $m/N^2 \ll 1$ we must take into account both the terms for which $K_p \gg 1$ and $K_p \ll 1$. We define the respective contributions to $P(n)$ coming from terms $K_p \gg 1$ and $K_p \ll 1$ as $P_L(n)$ and $P_S(n)$. Thus, $P(n) \approx P_L(n) + P_S(n)$.

The Case $K_p \gg 1$. Following Eq. (B.12), K_p can be large if $s = 0(1)$ and $\alpha_s \approx 1$ *or* s is large and $\mathrm{tg}(\alpha_s \pi/2) = O(1)$. In the first case ($s \ll K_p$) define y through $\alpha_s = 1 - y$, where $y \ll 1$. Hence

$$\mathrm{tg}(\alpha_s \pi/2) \approx \frac{1}{y\pi/2}$$

It follows from Eq. (B.12) that

$$y \approx \frac{2s+1}{K_p}, \qquad \alpha_s \approx 1 - \frac{2s+1}{K_p} \approx 1 - \frac{2s+1}{m \sin^2(\pi p/2N)} \tag{B.14}$$

Therefore

$$1 + \cos \alpha_s \pi \approx 2 \sin^2 \frac{2s+1)\,\pi}{2m \sin^2(\pi p/2N)} \approx \frac{\pi^2}{2m^2} \frac{(2s+1)^2}{\sin^4(\pi p/2N)} \tag{B.15}$$

In this case, expression (B.13) is approximately equal to

$$\frac{\pi^2}{m^3}\frac{(2s+1)^2}{\sin^4(\pi p/2N)}$$

In the second case (i.e., $s \gg 1$) it follows that [using expression (B.12)]

$$\operatorname{tg}(\alpha_s \pi) \approx \frac{2}{\pi^2}\frac{K_p}{s} \tag{B.16}$$

Since $s \gg 1$, by assumption, it follows that $\alpha_s \ll 1$ and expression (B.15) approximately equals

$$\frac{1}{m}\frac{1}{1+K_p^2/\pi^2 s^2}$$

Approximating $\cos(\theta/2)$, for $\theta \ll 1$, by an exponential and using Eqs. (B.8) and (B.11), we find for $s = O(1)$ ($\alpha_s \approx 1$)

$$\cos^{2n}(\theta/2) \approx \exp[(-\pi^2/4_m^2)(2s+1)^2\, n]$$

and for $s \gg 1$ ($\alpha_s \ll 1$)

$$\cos^{2n}(\theta/2) \approx \exp[(-\pi^2/m^2)\, s^2 n]$$

Finally, $P_L(n)$ reads

$$\begin{aligned} P_L(n) \approx{} & \frac{\pi^2}{m^3} \sum_{p=p_{\min}}^{N} (-)^{p+1} \frac{F(p)}{\sin^4(\pi p/2N)} \\ & \times \sum_{s=0}^{K_p} (2s+1)^2 \exp\left[-\frac{\pi^2}{4m^2}(2s+1)^2 n\right] \\ & + \frac{1}{m} \sum_{p=p_{\min}}^{N} (-)^{p+1} F(p) \sum_{s=K_p}^{m} \exp\left(-\frac{\pi^2}{m^2} s^2 n\right) \end{aligned} \tag{B.17}$$

where $p_{\min}$ is the smallest p for which $K_p \gg 1$. If $m/N^2 \gg 1$, then $p_{\min} = 1$ because $K_1 \gg 1$ in this case [see Eqs. (B.9) and (B.4)]

$$P_L(n) \approx P(n) \qquad \text{when} \quad m/N^2 \gg 1 \tag{B.18}$$

When $n \ll m^2$ the summation over s in the first term in Eq. (B.17) can be replaced by an integral over s. The contribution of this term, which we denote by $P_1(n)$, can be written

$$P_1(n) \approx \frac{1}{\sqrt{\pi}} \sum_{p=p_{\min}}^{N} (-)^{p+1} \frac{F(p)}{\sin^4(\pi p/2N)}\, n^{-3/2} \tag{B.19}$$

If also $n \gg N^4$, then the second term in Eq. (B.17) can be neglected with respect to $P_1(n)$, since

$$(K_{p_{\min}})^2 \frac{n}{m^2} > (K_1)^2 \frac{n}{m^2} \gg \frac{n}{N^4}$$

[see Eqs. (B.9) and (B.4)]. Hence, in this case $P_L(n) \approx P_1(n)$. As explained, this approximate equality is true for $m^2 \gg n \gg N^4$. In this case $P_L(n) \approx n^{-3/2}$ and we have a power law decay of $P_L(n)$.

Since the mean first passage time (MFT) is $O(mN^2)$, then, when $m/N^2 \gg 1$ [by Eq. (B.18)], $T_N(n)$ and $G_N(n)$ are within the range of power law behavior near the MFT. In practice, even if the second term is not exponentially small, one may still obtain a power law behavior dominating over the contribution of the second term of $P_L(n)$. When $N \gg m$, the second term is $O(1/m)$, whereas the first term in Eq. (B.17) is $O(N^4/m^3)$ (because of the sine in the denominator) and thus $P_L(n) \approx P_1(n)$ again. If $m \gg 1$ and $n \leqslant m^2$, the summation over s can be replaced by an integral as before and we obtain the same power law behavior. In this case the conditions are $N \gg m \gg 1$ and $N \ll n \ll m^2$, which implies $N \ll m^2 \ll N^2$.

We thus found two regimes in the (m, N, n) space where we expect power law behavior of $P_L(n)$. However, for $m \leqslant N^2$, one must add to $P_L(n)$ the terms with $K_p \ll 1$.

The Case $K_p \ll 1$. In the sequel we shall have to evaluate sums of the type

$$S = \sum_{p=1}^{N} (-)^p A\left(\frac{p}{N}\right) \tag{B.20}$$

where A is a function of p/N. To perform this sum, we define an index q such that $p = 2q + \sigma$, where $\sigma = 0, 1$. Equation (B.20) becomes

$$S = \frac{1}{N} \sum_{q=1}^{N/2} \frac{A(2q/N) - A(2q + 1/N)}{N^{-1}} \tag{B.21}$$

If $A(2q/N) - A((2q+1)/N)$ is $O(1/N)$,

$$S \approx \sum_{q=1}^{N/2} \left.\frac{dA}{dx}\right|_{x=2q/N} \frac{1}{N} \tag{B.22}$$

Approximating the sum by an integral, we find

$$S \approx \tfrac{1}{2}[A(1) - A(2/N)] \tag{B.23}$$

$K_p \ll 1$ implies $\alpha_s \ll 1$ from Eq. (B.12), which becomes

$$K_p \approx \tfrac{1}{2}\pi(2s + \alpha_s)(\alpha_s \pi/2) \tag{B.24}$$

Using Eq. (B.9), we have

$$\frac{m}{N^2} p^2 \approx \alpha_s(2s+\alpha_s)$$

Hence, for $s=0$

$$\alpha_0 \approx \frac{p}{N}\sqrt{m} \tag{B.25}$$

and for $s \neq 0$

$$\alpha_s \approx \frac{m}{N^2}\frac{p^2}{2s} \tag{B.26}$$

In Eq. (B.1) the terms with $K_p \ll 1$ (P_S) separate into two contributions: those for which $s=0$ and those for which $s \neq 0$. Thus

$$P_S(n) = P_{s=0} + P_{s\neq 0}$$

where

$$P_{s=0}(n) \approx \frac{1}{m} \sum_{p=1}^{N} (-)^{p+1} F(p) \exp\left(\frac{-\pi^2}{4mN^2} p^2 n\right) \tag{B.27}$$

From Eq. (B.13) the cosine factor to first order in α_s [i.e., keeping the term $O(m/N^2)$] is

$$\cos^{2n}\frac{\theta}{2} \approx \exp\left[\frac{-\pi^2}{m^2}\left(s^2+\frac{mp^2}{2N^2}\right)n\right]$$

Hence

$$P_{s\neq 0}(n) \approx \frac{1}{m} \sum_{p=1}^{N} \sum_{s\neq 0}^{m} (-)^{p+1} F(p) \exp\left[\frac{-\pi^2}{m^2}\left(s^2+\frac{mp^2}{2N^2}\right)n\right] \tag{B.28}$$

Obviously, the double summation in Eq. (B.28) decouples to a product of single summations. Hence

$$P_{s\neq 0}(n) \approx \frac{1}{m} \sum_{p=1}^{N} (-)^{p+1} F(p) \exp\left(\frac{-\pi^2}{2N^2 m} p^2 n\right) \sum_{s\neq 0}^{m} \exp\left(\frac{-\pi^2}{m^2} s^2 n\right) \tag{B.29}$$

which, approximating the sum over s by an integral (if $n < m^2$), becomes

$$P_{s\neq 0}(n) \approx \frac{1}{2(\pi n)^{1/2}} \sum_{p=1}^{N} (-)^{p+1} F(p) \exp\left(\frac{-\pi^2}{2mN^2} p^2 n\right) \tag{B.30}$$

If $n \geqslant m^2$, then the sum over s equals approximately $\exp[(-\pi^2/m^2)\,n]$. Note that if we write $P_{s=0}$ in the form

$$P_{s=0} = \frac{1}{m} f(n) \tag{B.31}$$

then $P_{s\neq 0}$ reads for $n < m^2$

$$P_{s\neq 0} = \frac{1}{2(\pi n)^{1/2}} f(2n) \tag{B.32}$$

and for $n > m^2$

$$P_{s\neq 0} = \frac{1}{m}\left[\exp\left(\frac{-\pi^2}{m^2} n\right)\right] f(2n) \tag{B.33}$$

We now calculate $f(n)$ for T_N and G_N separately. For T_N, by Eq. (B.3),

$$f(n) \approx \frac{\pi^2}{4N} \sum_{p=1}^{N} (-)^{p+1} \left(\frac{p}{N}\right)^2 \exp\left[\frac{-\pi^2}{4m}\left(\frac{p}{N}\right)^2 n\right]$$

which is of the form of Eq. (B.20), yielding, by Eq. (B.23),

$$f(n) \approx \frac{\pi^2}{2N^3} \exp\left(\frac{-\pi^2}{mN^2} n\right) \tag{B.34}$$

For G_N we use Eq. (B.5) and we find

$$f(n) \approx \frac{\pi}{N} \sum_{p=1}^{N} (-)^{p+1} \frac{p}{N} \exp\left[\frac{-\pi^2}{4m}\left(\frac{p}{N}\right)^2 n\right] \tag{B.35}$$

Using Eq. (B.23), we obtain

$$f(n) = \frac{\pi}{N^2} \exp\left(\frac{-\pi^2}{mN^2} n\right) \tag{B.36}$$

If $n \ll mN^2$, i.e., $n \ll$ MFT, the exponentials in Eq. (B.34) and (B.36) are approximately of order unity. We summarize the results of this Appendix:

a. When $m/N^2 \ll 1$, then, in the region $m^2 \ll n \ll mN^2$, $P(n)$ is approximately constant *(plateau region)*. The reason is that for $n \gg m^2$ the terms corresponding $K_p \gg 1$ are exponentially small [see Eq. (B.17)]. In this case the contribution $P_{s\neq 0}(n)$ to $P_S(n)$ is also multiplied by a factor $\sim e^{-n/m^2}$ [see Eq. (B.33)]. Therefore, only $P_{s=0}$ survives [see Eq. (B.31)]. Finally, for $n \ll mN^2$, $f(n)$ is approximately constant [see Eqs. (B.34), (B. 36)]. Compare this result with the plateau in Fig. 7.

b. When $m/N^2 \gg 1$ only terms corresponding to $K_p \gg 1$ contribute and we have an $n^{-3/2}$ behavior until $n \approx m^2$. Hence

$m/N^2 < 1$	$m^2 \ll n \ll mN^2$	*plateau*
$m/N^2 \gg 1$	$m^2 \gg n \gg mN^2$	$n^{-3/2}$

For n larger than $\max(m^2, mN^2)$ the decay is exponential.

B2. Asymptotic Behavior of $\Psi_m(n)$

$\Psi_m(n)$ is given by Eq. (A.20) and its pole equation is

$$1 = \sin\theta \operatorname{tg}(m\theta/2)$$

Hence, $C_p = 1$ and $K_p = m/2$ [see Eq. (B.9)] and in this case $K_p \gg 1$. By the previous analysis we have for $s \ll m/2$

$$\alpha_s \approx 1 - 2\frac{2s+1}{m}, \qquad \theta \approx \frac{\pi}{m}(2s+1)$$

For $s = O(m/2)$ we have $\alpha_s \approx m/\pi^2 s$ and $\theta \approx 2\pi s/m$ [i.e., $\theta = O(1)$], which is exponentially small for large n. Thus, Eq. (A.20) becomes

$$\Psi_m(n) \approx \frac{\pi^2}{m} \sum_{s=0}^{m} \left(\frac{2s+1}{m}\right)^2 \exp\left[\frac{-\pi^2}{4}\left(\frac{2s+1}{m}\right)^2 n\right] \tag{B.37}$$

Approximating the sum by an integral, we find

$$\Psi_m(n) \approx \frac{1}{\sqrt{\pi}} n^{-3/2} \tag{B.38}$$

which is valid as long as the exponential factor is $O(1)$, i.e., $n \ll m^2$, after which the decay is exponential (see Fig. 8). Thus, very long dangling bonds introduce an algebraic tail in the distribution functions.

APPENDIX C. PROBABILITY DISTRIBUTIONS WITH BIAS

As in the test, we have a probability p_1 to move from left to right on the backbone and $p_2 < p_1$ from right to left. We choose $p_1 + p_2 = 1/2$. This will not limit the generality of our results. As in the text [see Eq. (4.3)], one can develop a recursion relation for Q_N.[21] In principle one should define two types of Q_N, Q_N^+ and Q_N^-, corresponding to walks where the walker moves from point "0" back to point "0" or from point "N" back to point "*N*." However, it was shown in Ref. 19 that ${Q_N}^+ = {Q_N}^-$. We thus use Q_N to denote these quantities. Following Ref. 19,

$$Q_{N+1} = p_1 p_2 z^2 \frac{Y_m}{1 - Q_N Y_m} \tag{C.1}$$

where Y_m is defined in the text [see Eq. (4.2)].

It was also proven in Ref. 19 that

$$T_N^+ = \left(\frac{p_1}{p_2}\right)^N T_N^- \tag{C.2}$$

with obvious notation. The second recursion relation is

$$Q_{N+1} = Q_N + \frac{T_N^+ T_N^- Y_m}{1 - Y_m Q_N} \tag{C.3}$$

Using Eq. (C.2), we rewrite (C.3) to read

$$Q_{N+1} = Q_N + \left(\frac{p_2}{p_1}\right)^N \frac{T_N^{+2} Y_m}{1 - Y_m Q_N} \tag{C.4}$$

Define

$$\Lambda = \frac{1 - (1 - 4a)^{1/2}}{1 + (1 - 4a)^{1/2}} \tag{C.5}$$

and

$$a = p_1 p_2 z^2 Y_m^2 \tag{C.6}$$

Hence

$$Q_N = Y_m^{-1} \frac{\Lambda}{\Lambda + 1} \frac{1 - \Lambda^{N-1}}{1 - \Lambda^N} \tag{C.7}$$

and

$$T_N^+ = \left(\frac{p_1}{p_2}\right)^{N/2} (p_1 p_2)^{1/2} z \Lambda^{(N-1)/2} \frac{1 - \Lambda}{1 - \Lambda^N} \tag{C.8}$$

The poles of Q_N and T_N^+ are located at

$$\Lambda_p = e^{i2\pi p/N}, \qquad p = 1,..., N - 1 \tag{C.9}$$

Using Eqs. (C.5)–(C.6), we obtain

$$(p_1 p_2)^{1/2} z Y_m = {\Lambda_p}^{1/2} / (\Lambda_p + 1) \tag{C.10}$$

The "pole equation" is

$$(p_1 p_2)^{1/2} zY_m = \frac{1}{2\cos(\pi p/N)} \tag{C.11}$$

We can write G_N^+ as [see Eq. (A.15)]

$$G_N^+ = A_m T_N^+/(1 - A_m Q_N) \tag{C.12}$$

where

$$A_m^{-1} = Y_m^{-1} - p_2 z \tag{C.13}$$

or

$$G_N^+ = \left(\frac{p_1}{p_2}\right)^{N/2} (p_1 p_2)^{1/2} zY_m \frac{\Lambda^{(N-1)/2}(1-\Lambda^2)}{-[(p_2/p_1)\Lambda]^{1/2}(1-\Lambda^N) + 1 - \Lambda^{N+1}} \tag{C.14}$$

The poles in Eq. (C.14) must be found numerically in order to find the corresponding values of z. For each of these poles Eq. (C.10) must be solved (numerically). The residue must be calculated and the summation on the poles must be performed. In the limit of vanishing bias we of course recover Eq. (A.15). Another calculable limit is that of infinite bias, i.e., $p_1 = \frac{1}{2}$, $p_2 = 0$, i.e., the bias is so strong that the carrier has zero probability to jump against the field. In this case G_N^∞ (i.e., G_N for infinite bias) can be found directly as follows. Starting at site "0", the carrier performs any number of incursions into the loop, which yields Y_m, then goes to site "1," where the process repeats itself with $N-1$ sites left. Thus

$$G_N^\infty = \tfrac{1}{2} zY_m G_{N-1}^\infty = (\tfrac{1}{2} zY_m)^N \tag{C.15}$$

[in this case $G_N^\infty(z)$ obviously has multiple poles in the complex plane].

We return to the case of finite bias.

In order to calculate the MFT τ, it is useful to define a new variable ζ:

$$\Lambda = e^{2i\zeta} \tag{C.16}$$

G_N reads

$$G_N = \left(\frac{p_1}{p_2}\right)^{N/2} \frac{\sin\zeta}{\sin(N+1)\zeta - (p^2/p_1)^{1/2}\sin(N\zeta)} \tag{C.17}$$

The MFT is

$$\tau = (G_N'/G_N)_{z=1} \tag{C.18}$$

Define b, the bias parameter, as ($b \propto E$) (see Section 5)

$$p_1 = \kappa e^{b} \qquad \text{and} \qquad p_2 = \kappa e^{-b} \tag{C.19}$$

where

$$\kappa = 1/(4 \cosh b) \tag{C.20}$$

At $z = 1$ we have

$$\Lambda = \frac{1 - \text{th}\, b}{1 + \text{th}\, b} = e^{-2b} \tag{C.21}$$

$$\zeta = (-i/2) \ln \Lambda = ib \tag{C.22}$$

We finally obtain

$$\tau = (2m - 1) \coth b \left[\coth b - \frac{(N+1) \cosh[(N+1)\, b] - e^{-b} N \cosh(Nb)}{\sinh[(N+1)\, b] - e^{-b} \sinh(Nb)} \right] \tag{C.23}$$

For $bN \gg 1$ we have

$$\tau = N(2m - 1) \coth b \tag{C.24}$$

and for $b \to 0$ and $bN \ll 1$ we have, after some algebra,

$$\tau \approx N(N+1)(2m-1)(1 - bN)/3 \tag{C.25}$$

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