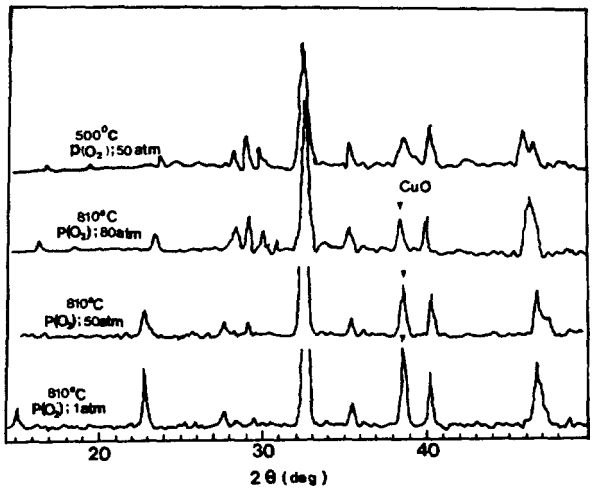

Figure 5. X-ray diffraction patterns of the 1-2-4 samples after sintering under the high oxygen pressure.

Cu_4O_8 superconductor by the EDTA complex pyrolysis method under 1 atm oxygen pressure. This method can overcome kinetic problems, since homogeneous fine powder is obtained. The starting material with 1-2-4 nominal composition was converted to the 1-2-3+CuO phase during densification, followed by the recovery of the 1-2-4 phase and the resultant samples were studied by the XRD, TG, DTA and AC magnetic susceptibility measurements. In conclusion, we have developed a new 1-2-4 synthesis requiring no special technique such as particularly high temperature and 1-2-4 phase is predominant as the $P(O_2)$ is increased.

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Average Walk Length in One-Dimensional Lattice Systems

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We consider the problem of a random walker on a one-dimensional lattice (N sites) confronting a centrally-located deep trap (trapping probability, $T=1$) and $N-1$ adjacent sites at each of which there is a nonzero probability s ($0<s<1$) of the walker being trapped. Exact analytic expressions for $<n>$ and the average number of steps required for trapping for arbitrary s are obtained for two types of finite boundary conditions (confining and reflecting) and for the infinite periodic chain. For the latter case of boundary condition, Montroll's exact result is recovered when s is set to zero.

Introduction

The theories of random walks on a space lattice have been dealt for a couple of decades since the turn of the century[1-4]. There are numerous examples which can be explained by a random walk on a space lattice. To name some of them, diffusion of electrons, excitons, energy transfer[3,5,6], etc.

In this paper for a particular class of one-dimensional lattice problem, we have shown an analytic expression for the average walk length on a chain with a centrally-disposed

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deep trap (trapping probability, $T=1$) flanked by $N-1$ sites at which the probability of being trapped is uniformly non-zero ($0<s<1$), for two types of finite boundary conditions (confining (or transmitting) and reflecting) and for infinite periodic chain.

We develop in the following section recursion relations from which analytic expressions for $<n>$ are constructed for $T=1$, arbitrary s and for both finite and infinite boundary conditions.

In section III we consider the special case, $T=1$ with all $s=0$, for the particular case of periodic boundary condition. We show how the classic result ($<n>=\frac{N(N+1)}{6}$, $N=2N+1$)[1-3] of Montroll may be recovered from our theory and then present a companion result for the same trapping problem but for the case of reflecting boundary conditions. As it happens, the details of the general proof presented in section II do not depend on the assumption that the trapping probability s of the $N-1$ sites adjacent to the central deep trap be constant. Rather, s can be chosen to depend parametrically on some auxiliary variable of the problem. For example, in problems dealing with the catalytic effectiveness of linear supports or in the problem of exciton migration, the time is a relevant variable of the process being considered[5,6].

Accordingly we explore in section IV the consequences of assuming two different functional forms describing the dependence of the trapping probability s on the temporal state of the system. In particular, a linear and an exponential dependence of s on the (reduced) time T is postulated and profiles of $<n>$ versus T for each case are presented.

Theory for Arbitrary *s*.

Periodic and Confining Boundary Conditions. Let us construct a generating function $U_i(t)$ for the trapping probability distribution associated with lattice site i. It is

$$U_i(t)=\sum_{n=0}^{\infty} t^n P_i(n),$$

where $P_i(n)$ is the probability that an rendom walker starting from site i becomes trapped at a centrosymmetric trap after n^{th} step.

By standard manipulations, we get

$$U_i(t)-t\sum_{i'} p(i-i')U_{i'}(t)=U_i(0) \tag{1}$$

where $p(i-i')$ is the transition probability from site i to site $i'(i\neq i')$. If $p(i-i')$ is independent of n, *i.e.*, we are dealing with a Markovian process, then the above expression (1) can be expressed as a vector equation in which the ith element represents the generating function of site i.

If we write (1) in matrix form,

$$\bar{U}(t)-tP\bar{U}(t)=\bar{U}(0) \tag{2}$$

where $\bar{U}(0)$ is the initial condition of the problem and P is the transition probability matrix.

From now on, we denote matrix as $P(N)$ and determinant as P_N.

Inverting Eq. (3), we have

$$\bar{U}(t)=(1-tP)^{-1}=\bar{U}(0) \tag{3}$$

Sinece the matrix $(1-tP)$ can be specified at the outset, it is seen that the quantity $\bar{U}(t)$ can be evaluated via a simple inversion of the matrix $(1-tP)$. Once this is achieved, the average walk length vector $<\bar{n}>$ can be derived from

$$[\bar{n}]=\frac{\partial\bar{U}(t)}{\partial t}\Big|_{t=1}=\frac{\partial}{\partial t}[(1-tP)\bar{U}(0)]\Big|_{t=1} \tag{4}$$

or,

$$<\bar{n}>=\frac{P}{1-P}\bar{U}(1) \tag{5}$$

Consider now the particular case of a random walk on a one-dimensional lattice. We assume that there exists a centrosymmetric deep trap (T) flanked by N sites at each of which there exists a finite probability s of being trapped. We impose periodic boundary conditions on the resultant $2N+1=N$ site, odd lattice. If hopping to nearest neighbor sites only is permitted then an $(N+1)\times(N+1)$ transformation matrix can be identified as

$$\begin{bmatrix} 1 & 0 & 0 & \cdots & \\ \alpha & 1 & \alpha & 0 & \cdots \\ 0 & \alpha & 1 & \alpha & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots & \alpha \\ 0 & 0 & 0 & 0 & \cdots 1+\alpha \end{bmatrix} \begin{bmatrix} <n>_T \\ <n>_1 \\ <n>_2 \\ \cdot \\ \cdot \\ <n>_N \end{bmatrix}$$

$$=\begin{bmatrix} 0 \\ -\alpha\{U_1(1)+U_3(1)\} \\ -\alpha\{U_1(1)+U_4(1)\} \\ \cdots \\ -\alpha\{U_{N-1}(1)+U_{N+1}(1)\} \\ -\alpha\{U_N(1)+U_{N+1}(1)\} \end{bmatrix} \tag{6}$$

where, $\alpha=-\frac{1-S}{2}$.

It can be easily seen that if $s=0$, $\bar{U}(1)\equiv\begin{bmatrix}1\\1\\\vdots\\1\end{bmatrix}$,

from Eq. (3) and initial condition $\bar{U}(0)=\begin{bmatrix}1\\0\\\vdots\\0\end{bmatrix}$

For $s\neq 0$, $\bar{U}(1)\equiv\frac{1}{Q_N}\begin{bmatrix} Q_N \\ (-\alpha)Q_{N-1} \\ (-\alpha)^2Q_{N-2} \\ \vdots \\ (-\alpha)^NQ_0 \end{bmatrix}$

Therefore (6) turns out to be (ref. Eq. (11) and (14))

$$\begin{bmatrix} 1 & 0 & 0 & \cdots & & 0 \\ \alpha & 1 & \alpha & 0 & \cdots & 0 \\ 0 & \alpha & 1 & \alpha & \cdots & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ 0 & 0 & 0 & 0 & \cdots \alpha & 1+\alpha \end{bmatrix} \begin{bmatrix} <n>_T \\ <n>_1 \\ <n>_2 \\ \cdot \\ \cdot \\ <n>_N \end{bmatrix}$$

$$=\frac{1}{Q_N}\begin{bmatrix} Q_N \\ (-\alpha)Q_{N-1} \\ (-\alpha)^2Q_{N-2} \\ \vdots \\ (-\alpha)^NQ_0 \end{bmatrix} \tag{7}$$

Therefore the problem of determining $\langle\bar{n}\rangle$ is reduced to the problem of inverting the matrix $Q(N+1)$.

To compute the inverse of $Q(N+1)$ we must determine the adjoint matrix and obtain an expression for det $Q(N+1)$. Speaking to the latter problem first, let us define the anxiliary quantity[7],

$$R_N=\det\begin{bmatrix} 1 & \alpha & 0 & 0 & 0 & \cdots & 0 \\ \alpha & 1 & \alpha & 0 & 0 & \cdots & 0 \\ 0 & \alpha & 1 & \alpha & 0 & \cdots & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ 0 & 0 & 0 & & & 1 & \alpha \\ 0 & 0 & 0 & & & \alpha & 1 \end{bmatrix} \tag{8}$$

Using this, the determinant of the matrix $Q(N+1)$ may be written,

$$\det\ Q(N+1)=(1+\alpha)R_{N-1}-\alpha^2R_{N-2}=Q_N \tag{9}$$

The R_N can be expressed as a recurrence relation of following the form.

$R_N=R_{N-1}-\alpha^2R_{N-2}$ with initial values : $R_1=1$ and $R_2=1-\alpha^2$. We define generating function Z,

$$Z=1+R_1X+R_2X^2+R_3X^3+\cdots$$

which, taking account of the initial values R_1 and R_2, gives the closed form expression :

$$Z=\frac{1}{1-X+\alpha^2X^2} \tag{9}$$

It is convenient to specify the range of values of the parameter α by an angular variable θ as $\alpha=\frac{1}{2}\cos\theta\left(\frac{\pi}{2}\leq\theta<\pi\right)$. Then one obtains the result

$$R_N=\left(\frac{1}{\sin\theta}\right)\left[\frac{(1+\sin\theta)^{N+1}}{2^{N+1}}-\frac{(1-\sin\theta)^{N+1}}{2^{N+1}}\right] \tag{10}$$

by equating corresponding terms in the two expressions for Z. This procedure for determining R_N does not work for the special case $s=0$, however, one can show, either *via* application of L' Hospital's rule or *via* direct Taylor expansion, that $R_N=\frac{(N+1)}{2^N}$ for $s=0$.

Nest, we determine the adjoint matrix corresponding to $Q(N+1)$.

The cofactor of each element of the maxtric $Q(N+1)$ may be worked out with the results :

$$A_{11}=Q_N \tag{11a}$$

$$A_{j1}=0 \text{ for } j=2 \text{ thru } j=N+1 \tag{11b}$$

$$A_{1j}=(-1)^{1+j}\alpha^{j-1}Q_{N-j+1} \text{ for } j=2 \text{ thru } j=N+1 \tag{11c}$$

$$A_{ij}=(-1)^{i+j}\alpha^{i-j}R_{j-2}Q_{N-i+1} \text{ for } i\geq j \text{ and } i=2 \text{ thru } i=N+1, \tag{11d}$$

and finally,

$$A_{ij}=(-1)^{i+j}\alpha^{i-j}R_{i-2}Q_{N-j+1} \text{ for } i<j \text{ and } j=2 \text{ thru } j=N+1, \tag{11e}$$

with

$$Q_0=1$$

$$Q_1=1+\alpha$$

$$Q_2=1+\alpha-\alpha^2.$$

Either Eq. (6) or (7) can be used for calculation of the average walk length $\langle n\rangle$ from site j.

From Eq. (7)

$$\langle n\rangle_{j-1}=\frac{1}{Q_N^2}\sum_{i=2}^{N+1}A_{ij}(-\alpha)^{j-1}Q_{N-i+1} \tag{12}$$

where the index j ranges from 2 to $N+1$. Using this expression in conjunction with the earlier results, Eqs. (8), (10) and (11) allows the calculation of the average walk length from an arbitrary site j of the one-dimensional lattice.

In order to express more clearly the analytic structure of the result, we enter Eqs. (8), (10) and (11) into the Eq. (12), with the result :

$$\langle n\rangle_{j-1}=\frac{1}{Q_N^2}\left[(-\alpha)^{j-1}Q_{N-j+1}\sum_{i=2}^{j}R_{i-2}Q_{N-i+1}+(-\alpha)^{-j-1}R_{j-2}\sum_{i=j+1}^{N+1}\{(-\alpha)^iQ_{N-i+1}\}^2\right] \tag{13}$$

where,

$$Q_{N-j+1}=\frac{1}{2\sin\theta}\left\{\left(\frac{1+\sin\theta}{2}\right)^{N-j+1}\left(1+\sqrt{2}\sin\frac{2\theta+\frac{\pi}{2}}{2}\right)-\left(\frac{1-\sin\theta}{2}\right)^{N-j+1}\left(1-\sqrt{2}\sin\frac{2\theta-\frac{\pi}{2}}{2}\right)\right\} \tag{14}$$

The use of Eqs. (10), (13) and (14) allows the explicit calculation of $\langle\bar{n}\rangle$ for arbitrary s, and from these one can determine the overall walk length $\langle n\rangle$:

$$\langle n\rangle=\frac{\sum_{i=1}^{N}\sigma_i\langle n\rangle_i}{2N}=\frac{1}{N}\sum_{i=1}^{N}\langle n\rangle_i \tag{15}$$

where the symmetry number σ for the one-dimensional lattice here is 2 for all sites i.

In concluding this subsection, we comment on the relationship between the results obtained for periodic boundary conditions versus those obtained for the case of confining boundary conditions. By the latter class of boundary conditions is meant that if the walker attempts to step off the end (or Nth) site of the one-dimensional lattice, he must return to the lattice site from which he started (*i.e.*, the Nth site). It turns out that the mathematical structure of the transformation matrix for the latter problem is exactly the same as that for the infinite periodic chain.

Therefore, the analytic results reported above (*viz.*, Eqs. (13), (14) and (15)) apply as well to a finite, one-dimensional chain with a centrosymmetic deep trap.

Reflecting Boundary Conditions. The case of reflecting boundary conditions is implemented by the restriction that if the walker attempts to step off the end (or Nth) site of the lattice, he is displaced to one (interior) lattice point further from thee boundary than the lattice site from where he started.

In this case, if hopping to nearest neighbors only is permitted, then an $(N\times1)\times(N\times1)$ transformation matrix descriptive of the process can be written down and Eq. (4) assumes the form :

$$\begin{bmatrix} 1 & 0 & 0 & 0 & \cdots & \cdots \\ \alpha & 1 & \alpha & 0 & \cdots & \cdots \\ \cdots & & & & & \\ \cdots & & & & & \\ \cdots & & \alpha & 1 & \alpha & \\ \cdots & & 0 & (2\alpha) & 1 & \end{bmatrix} \begin{bmatrix} <n>_T \\ <n>_1 \\ <n>_2 \\ \cdot \\ \cdot \\ <n>_N \end{bmatrix}$$

$$= \frac{1}{Q_N} \begin{bmatrix} 0 \\ (-\alpha)Q_{N-2} \\ (-\alpha)^2 Q_{N-1} \\ \cdot \\ (-\alpha)^j Q_{N-j} \\ \cdot \\ 2(-\alpha)^N Q_1 \end{bmatrix} \tag{16}$$

where here, $\alpha = -\frac{1-S}{2}$

If we define the transformation matrix on the left-hand side as $Q(N+1)$, then with the auxiliary quantity R_N, we find that $Q_N = \det Q(N) = R_{N-1} - 2\alpha^2 R_{N-2}$, with in this case, $Q_0 = 1$, $Q_1 = 1$, $Q_2 = 1 - 2\alpha^2, \cdots$

Following a procedure exactly similar to that laid down in the preceding section, we invert the matrix Eq. (16) and find,

$$<n>_{j-1} = \frac{1}{Q_N^2} \sum_{i=2}^{N+1} A_{ij}(-\alpha)^{i-1} Q_{N-i-1}, \tag{17}$$

where, in this case,

$$Q_{N-i+1} = \left(\frac{1+\sin\theta}{2}\right)^{N-i+1} + \left(\frac{1-\sin\theta}{2}\right)^{N-i+1}$$

This result may be used in conjunction with Eq. (15) to determine the overall walk length $<\bar{n}>$ for a one-dimensional lattice with a centrosymmetric deep trap and reflecting boundary conditions. Representative calculations of $<\bar{n}>$ for arbitrary s for the three boundary conditions studied in this paper will be presented in section IV.

The Special Case. $s=0$

The choice $s=0$ together with periodic boundary conditions corresponds to the problem considered a generation ago by Montroll and Weiss for which it was proved that,

$$<n> = \frac{N(N+1)}{6}, \tag{18}$$

where N is the total number of lattice sites on the chain.

The result (18) can be recovered either from the expressions obtained from matrix Eq. (7), or from matrix Eq. (6).

To generate $<n>$ starting from Eq. (6), we need two preliminary results.

First, from the expression (10) we have $(t=1)$:

$$\lim_{s\to 0} R_N = \lim_{s\to 0}\left\{\frac{1}{\sin\theta}\left[\frac{(1+\sin\theta)^{N+1}}{2^{N+1}} - \frac{(1-\sin\theta)^{N+1}}{2^{N+1}}\right]\right\} = \frac{N+1}{2^N}$$

Next, we have $\frac{\partial R_N}{\partial t} = \sum_{i=1}^{N} \det R'_i(N)$ evaluated at $t=1$ and $s=0$, where $R'_i(N)$ is the $N\times N$ matrix:

$$R'_i(N) = \begin{bmatrix} 1 & \alpha & 0 & 0 & \cdots & \cdots \\ \alpha & 1 & \alpha & 0 & \cdots & \cdots \\ \cdots & & & & & \\ \cdots & & & & & \\ \cdots & & \frac{\partial\alpha}{\partial t} & 0 & \frac{\partial\alpha}{\partial t} & \\ \cdots & & \alpha & 1 & \alpha & \\ \cdots & & & \alpha & 1 & \end{bmatrix} \tag{20}$$

where, $\alpha = -\frac{1}{2}t$

Then we obtain the following recursion relation for $R(N)$:

$$R'_i(N) = (-1)^{2i-1}\frac{\partial\alpha}{\partial t}\Big|_{t=1} \alpha R_{i-2}R_{N-i} + (-1)^{2i-1}\frac{\partial\alpha}{\partial t}\Big|_{t=1} \alpha R_{i-1}R_{N-i-1} \tag{21}$$

From Eqs. (20) and (21),

$$\frac{dR_N}{dt}\Big|_{t=1} = (-1)\left(\alpha\frac{\partial\alpha}{\partial t}\right)\Big|_{t=1}\left\{\sum_{i=1}^{N}(R_{i-2}R_{N-i} + R_{i-1}R_{N-i-1})\right\}\Big|_{t=1} \tag{22}$$

From above, we have immediately:

$$Q_N|_{t=1} = [(1+\alpha)R_{N-1} - \alpha^2 R_{N-2}]|_{t=1} = \frac{1}{2^N},$$

and

$$\frac{\partial Q_N}{\partial t}\Big|_{t=1} = -\frac{N^2}{2^N},$$

with these results, the average walk lenght from site i to the centrosymmetric trap may be calculated by differentiating the generating function directly.

$$<n>_{j-1} = \frac{\partial U_{j-1}}{\partial t}\Big|_{t=1} = \frac{\partial}{\partial t}\left(\frac{A_{ij}}{Q_N}\right)\Big|_{t=1}$$

$$= -j^2 + j(2N+3) - 2(N+1) \tag{23}$$

A similar calculation starting from Eq. (4) also gives the $<\bar{n}>$,

$$<n>_{j-1} = \frac{1}{Q_N}\sum_{i=1}^{j} A_{ij} + \sum_{i=j+1}^{N+1} A_{ij}$$

Using the previous results (for Q_N and R_N) we have

$$<n>_{j-1} = -j^2 + j(2N+3) - 2(N+1). \tag{24}$$

Therefore, Eqs. (23) and (24) give the overall average walk length to be,

$$<n> = \frac{1}{N}\sum_{j=2}^{N+1} <n>_{j-1} = \frac{(2N+2)(2N+1)}{6} = \frac{N(N+1)}{6}$$

(Notice that in our notation Montroll's N corresponds to $2N+1$).

For the case of reflecting boundary conditions,

$$<n>_{j-1} = -j^2 + 2j(N+1) - (2N+1)$$

Consequently, the overall walk length $<n>$ is

$$<n> = \frac{1}{N}\sum_{j=2}^{N+1} <n>_{j-1} = \frac{(N+1)(4N-1)}{6} = \frac{(N+1)(2N-3)}{12}$$

Discussion

The special case $s=s(t)$. Numerical results.

Assigning s to be nonzero value bounded between zero and unity means that the background sites are no longer

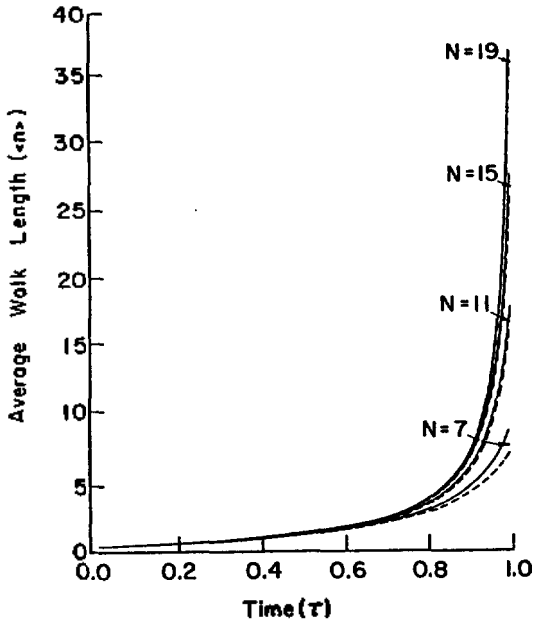

Figure 1. Evaluation profiles fot the overall expected walk length $<n>$ versus reduced time T assuming that $s=1-T$. Each pair of curves relates to a lattice with N sites characterized by a trapping probability s on either side of a central, deep trap ($T=1$) and subject to periodic (solid line) or reflecting (dotted line) boundary conditions.

nonabsorbing but rather can, with probability s, trap the walker.

In the spirit of reaction-diffusion theory, if we regard the random walker as a molecule (atom, exciton) diffusing toward a target molecule (at the deep trap), then setting $s=0$ means that the $N-1$ sites flanking the target molecule can be regraded as reaction centers which with nonvanishing probability s, may react with the diffusing species, thereby removing the coreactant irreversibly from the system.

In fact, we may imagine following physical situations, where the probability s is time dependent. For example, consider a situation where all the sites are characterized initially by $s=1$. Then with the passage of time we may imagine that the $N-1$ auxialiary sites except the central trap become systematically and uniformly depressed. In this situation, one would anticipates s as a function of time. Then in this situation, we may proceed by imposing a functional dependence on s in terms of a dimensionless time variable ($T=t/t_c$, where t_c is some characteristic lifetime). Two sorts of functional dependences are considered:

$$s(T)=1-T \tag{25}$$

and

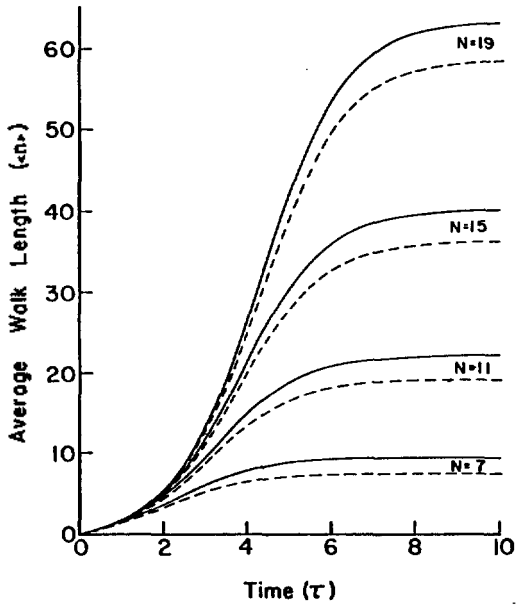

Figure 2. Evaluation profiles for the overall expected walk length $<n>$ versus reduced time T assuming that $x=e^{-T}$. The conventions here are the same as in Figure 1.

$$s(T)=e-T \tag{26}$$

The results for the first case for periodic and reflecting boundary conditions for a series of one-dimensional lattices: $N=7$, 11, 15, 19, are shown in Figure (1).

It is seen that a linear response to the external environment leads to an initial, slowly varing change in the effeciencyy of reaction of the system, with a dramatic rise to the limiting value $<n>_{s=0}$ realized only when the $N-1$ background sites have been essentially neutralized. The exponential decay in the reaction effeciency of the $N-1$ auxiliary sites specified by the Eq. (25) leads to short-time behavior similar to that given by Eq. (26).

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