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Multiple Trapping of Random Walkers on Periodic Lattices

Alberto Robledo[1] and Luis Woodhouse[1]

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Formulas are obtained for the mean absorption time of a set of k independent random walkers on periodic space lattices containing q traps. We consider both discrete- (here we assume simultaneous stepping) and continuous-time random walks, and find that the mean lifetime of the set of walkers can be obtained, via a convolution-type recursion formula, from the generating function for one walker on the perfect lattice. An analytical solution is given for symmetric walks with nearest neighbor transitions on N-site rings containing one trap (or q equally spaced traps), for both discrete and exponential distribution of stepping times. It is shown that, as $N \to \infty$, the lifetime of the walkers is of the form Ta_kN^2, where T is the average time between steps. Values of a_k, $2 \leqslant k \leqslant 6$, are provided.

KEY WORDS: Multiple trapping; mean absorption time; lattice random walks.

1. INTRODUCTION

An important problem in random walk theory is the evaluation of the average number of steps that a walker on a periodic space lattice containing traps at some preassigned sites requires to take before trapping. This problem arises, for instance, when some physical or biological phenomena that involve the transport of excitation energy through a network of molecules to specialized centers are modeled by random walks. Examples of these are luminescent emission from a polymer[(1)] or an organic crystal[(2)] and primary processes in photosynthesis.[(3,4)]

Assuming that a walker has the same probability of starting from any nontrapping site, Montroll[(5)] obtained exact analytical results for the mean lifetime of one walker, for random walks on lattices with periodic distribu-

[1] División de Estudios Superiores, Facultad de Química, Universidad Nacional Autónoma de México, Mexico.

tions of traps, or, equivalently, on finite lattices with periodic boundary conditions containing only one trap. His method, which employs generating function techniques, constituted an alternative approach to previous machine calculations[6] for the same process. Subsequently, this method has been applied to study the effect on trapping times due to lattice vibrations,[7] non-nearest-neighbor transitions,[8] and random distributions of traps.[9]

Here, we employ the generating function technique to study the mean absorption time of a set of random walkers. That is, we consider the successive trapping of a number of walkers on a finite lattice where some sites act as irreversible traps. The motivation of this work is a recent model for the energy trapping center in a photosynthetic unit proposed by Fong.[10] As a main feature, this model requires the arrival of two excitons at the active center before the chemical reaction can be triggered. It is our aim to investigate the duration of the kind of many walker process implied by this model. The emphasis in this paper is on development of techniques and methodology rather than on applications.

We shall introduce a formalism which is applicable to any number of walkers on a finite, d-dimensional lattice with periodic boundary conditions (d-torus) and which contains traps at fixed points. We shall assume that: (i) all walkers have the same probability of starting from any nontrapping point, and (ii) the walkers are independent, that is, they do not interfere with each other, thus allowing multiple occupancy of a single site at a given time. We find it convenient to refer first, in Section 2, to discrete-time random walks, and thus we shall further assume there that (iii) the walkers step at the same times and these steps occur at fixed time intervals. In Section 3, we relax this last assumption and derive multiple-trapping formulas for continuous-time variable, whereby walkers step according to a common but otherwise arbitrary probability density. Subsequently, in Section 4, we present a detailed application for k walkers on one-dimensional rings containing one trap (or q equally spaced traps), each walker with an equal probability to reach any nearest neighboring site on a given step. We consider two cases: stepping at regular time intervals and exponentially distributed stepping times.

2. GENERAL FORMALISM

We shall briefly review,[9] in a form suitable for generalization, the formalism for the trapping of one walker by q traps, before proceeding to the general case of k walkers in the presence of q traps.

Let us consider a d-dimensional periodic lattice containing N sites and suppose that (irreversible) traps exist at the set of q points $Q = \{\mathbf{s}_1, \mathbf{s}_2, \ldots, \mathbf{s}_q\}$. Let $F_n(\mathbf{s}_0)$ be the probability that a walker, which starts to walk at site $\mathbf{s}_0 \notin Q$, is trapped at the nth step, and let us define the generating function of the set $\{F_n(\mathbf{s}_0)\}$ as

$$F(\mathbf{s}_0, z) = \sum_{n=1}^{\infty} F_n(\mathbf{s}_0) z^n \tag{1}$$

Then, if a walker has the same probability of being at any nontrapping point on the lattice at the beginning of his walk, the average number of steps required to be trapped is given by

$$\langle n \rangle^{(1,q)} = \frac{1}{N-q} \sum_{\mathbf{s}_0 \notin Q} \frac{\partial}{\partial z} F(\mathbf{s}_0; z) \bigg|_{z=1} \tag{2}$$

Now, if we introduce the expression

$$R_n^{(1,q)} = \sum_{\mathbf{s}_0 \notin Q} \sum_{j=1}^{n} F_j(\mathbf{s}_0), \qquad R_0^{(1,q)} = 0 \tag{3}$$

we have that

$$R_n^{(1,q)} - R_{n-1}^{(1,q)} = \sum_{\mathbf{s}_0 \notin Q} F_n(\mathbf{s}_0) \tag{4}$$

and thus, in terms of the generating function of the $R_n^{(1,q)}$,

$$R^{(1,q)}(z) = \sum_{n=1}^{\infty} R_n^{(1,q)} z^n \tag{5}$$

Eq. (2) is expressed as

$$\langle n \rangle^{(1,q)} = \frac{1}{N-q} \frac{d}{dz} (1-z) R^{(1,q)}(z) \bigg|_{z=1} \tag{6}$$

The function $R^{(1,q)}(z)$ can be related to the generating function $P(\mathbf{s}; z)$ for walks which start at the origin on the perfect lattice (i.e., without traps). This function is in turn obtained in terms of the basic function $p(\mathbf{s})$ that specifies the detailed nature of the walk. $p(\mathbf{s})$ is the probability that any step results in a displacement $\mathbf{s}$ by a walker. Briefly, it can be shown[11] that, on a lattice with periodic boundary conditions,

$$P(\mathbf{s}; z) = N^{-1} \sum_{k_1=0}^{m-1} \cdots \sum_{k_d=0}^{m-1} [\exp(2\pi i \mathbf{s} \cdot \mathbf{k}/m)][1 - z\lambda(2\pi \mathbf{k}/m)]^{-1} \tag{7}$$

where $m^d = N$ and where $\lambda(\mathbf{\Theta})$, the structure function for the walk, is the Fourier expansion of $p(\mathbf{s})$, i.e.,

$$\lambda(\mathbf{\Theta}) = \sum_{\mathbf{s}} p(\mathbf{s}) \exp(i\mathbf{\Theta} \cdot \mathbf{s}) \tag{8}$$

$P(\mathbf{s}; z)$ generates the numbers $\{P_n(\mathbf{s})\}$, where $P_n(\mathbf{s})$ is the probability that a walker, starting at the origin, reaches site $\mathbf{s}$ at the nth step, independently of how many previous visits he has already had at $\mathbf{s}$.

In order to relate $R^{(1,q)}(z)$ to $P(\mathbf{s}; z)$, we first write $F(\mathbf{s}_0; z)$ in the form

$$F(\mathbf{s}_0; z) = \sum_{l=1}^{q} f(\mathbf{s}_l - \mathbf{s}_0; z) \tag{9}$$

where

$$f(\mathbf{s}_l - \mathbf{s}_0; z) = \sum_{n=1}^{\infty} f_n(\mathbf{s}_l - \mathbf{s}_0)z^n \tag{10}$$

and where $f_n(\mathbf{s}_l - \mathbf{s}_0)$ is the probability that a walker, on the perfect lattice, starting from $\mathbf{s}_0$ and avoiding the sites $\mathbf{s}_m \in Q$, $m \neq l$, reaches site $\mathbf{s}_l$ for the first time. The $\{f_n(\mathbf{s}_l - \mathbf{s}_0)\}$ satisfy the set of equations

$$\sum_{m=1}^{q} \sum_{k_m=1}^{n} P_{n-k_m}(\mathbf{s}_l - \mathbf{s}_m) f_{k_m}(\mathbf{s}_m - \mathbf{s}_0) = P_n(\mathbf{s}_l - \mathbf{s}_0), \qquad 1 \leqslant l \leqslant q, \quad \mathbf{s}_0 \notin Q \tag{11}$$

Equations (11) merely state the fact that the family of n-step walks between $\mathbf{s}_0$ and $\mathbf{s}_l$ $(1 \leqslant l \leqslant q)$ that give rise to $P_n(\mathbf{s}_l - \mathbf{s}_0)$ can be separated into different groups according to which of the sites $\mathbf{s}_m \in Q$ is visited first. By multiplying (11) by z^n and then summing over all n, we find

$$\sum_{m=1}^{q} P(\mathbf{s}_l - \mathbf{s}_m; z) f(\mathbf{s}_m - \mathbf{s}_0; z) = P(\mathbf{s}_l - \mathbf{s}_0; z), \qquad 1 \leqslant l \leqslant q \tag{12}$$

Finally, by taking into account Eq. (9), and noting that Eqs. (3) and (5) imply

$$R^{(1,q)}(z) = \frac{1}{1 - z} \sum_{\mathbf{s}_0 \notin Q} F(\mathbf{s}_0; z) \tag{13}$$

the resolution of the linear system (12) for the $f(\mathbf{s}_m - \mathbf{s}_0; z)$ yields the desired expression for $R^{(1,q)}(z)$ in terms of the basic function $P(\mathbf{s}; z)$. In particular, for the special cases of one and two traps, we obtain, respectively

$$R^{(1,1)}(z) = \frac{1}{(1 - z)^2 P(\mathbf{0}; z)} - \frac{1}{1 - z} \tag{14}$$

and

$$R^{(1,2)}(z) = \frac{2}{(1 - z)^2[P(\mathbf{0}; z) + P(\mathbf{s}_2 - \mathbf{s}_1; z)]} - \frac{2}{1 - z} \tag{15}$$

In arriving at Eqs. (14) and (15), we have employed the conservation condition

$$\sum_{\mathbf{s}} P(\mathbf{s}; z) = (1 - z)^{-1} \tag{16}$$

With regard to the significance of the numbers $R_n^{(1,q)}$, it is interesting to note that $R_n^{(1,1)}$ is the average number of distinct sites visited in an n-step walk (not including the starting point). More elaborate definitions in terms of visited sites can be assigned to $R_n^{(1q)}$ for $q > 1$.

k Walkers and q Traps

We consider next two independent walkers on the lattice that step at the same times and with initial positions at sites $\mathbf{r}_0$ and $\mathbf{s}_0$, neither of these a trap. The probability for one walker to be trapped at the nth step, conditioned to the other walker being trapped during this n-step walk, is given by

$$G_n(\mathbf{r}_0, \mathbf{s}_0) = F_n(\mathbf{r}_0) \sum_{j=1}^{n} F_j(\mathbf{s}_0) + F_n(\mathbf{s}_0) \sum_{j=1}^{n-1} F_j(\mathbf{r}_0) \tag{17}$$

Then, the average number of steps required for the two walkers to be trapped, independently of what their initial positions were, is

$$\langle n \rangle^{(2,q)} = \frac{1}{(N-q)^2} \sum_{\mathbf{r}_0, \mathbf{s}_0 \notin Q} \frac{\partial}{\partial z} G(\mathbf{r}_0, \mathbf{s}_0; z) \bigg|_{z=1} \tag{18}$$

where

$$G(\mathbf{r}_0, \mathbf{s}_0; z) = \sum_{n=1}^{\infty} G_n(\mathbf{r}_0, \mathbf{s}_0) z^n \tag{19}$$

In analogy with the one-walker case, the employment of Eqs. (3) and (4) in Eq. (17) permits the mean lifetime of the two walkers, Eq. (18), to be expressed as

$$\langle n \rangle^{(2,q)} = \frac{1}{(N-q)^2} \frac{d}{dz} (1 - z) R^{(2,q)}(z) \bigg|_{z=1} \tag{20}$$

where $R^{(2,q)}(z)$ is the generating function for the square of $R_n^{(1,q)}$, i.e.,

$$R^{(2,q)}(z) = \sum_{n=1}^{\infty} [R_n^{(1,q)}]^2 z^n \tag{21}$$

This function can be evaluated from the one-walker function $R^{(1,q)}(z)$ invoking the convolution property of the z-transform. From Eqs. (5) and (21), we obtain the relation

$$R^{(2,q)}(z) = \frac{1}{2\pi i} \int_{\Gamma_0} R^{(1,q)}(z'^{-1}) R^{(1,q)}(zz') z'^{-1} \, dz' \tag{22}$$

where the contour of integration is the unit circle $|z| = 1$.

By following the same procedure we have just outlined for the one- and two-walker cases, we can easily show that the mean absorption time for a set of k walkers, stepping at the same times, is given by

$$\langle n \rangle^{(k,q)} = \frac{1}{(N-q)^k} \frac{d}{dz} (1 - z) R^{(k,q)}(z) \bigg|_{z=1} \tag{23}$$

where

$$R^{(k,q)}(z) = \sum_{n=1}^{\infty} [R_n^{(1,q)}]^k z^n \tag{24}$$

We omit details of the derivation of Eq. (23) to avoid cumbersome notation and near repetition.

Similarly, the k-walker function $R^{(k,q)}(z)$ can be related to its lower order counterparts by means of the recursion relation

$$R^{(k,q)}(z) = \frac{1}{2\pi i}\int_{\Gamma_0} R^{(1,q)}(z'^{-1})R^{(k-1,q)}(zz')z'^{-1}\,dz' \tag{25}$$

thus reducing the problem of the evaluation of $\langle n\rangle^{(k,q)}$ to the repeated use of Eq. (25), provided the one-walker function $R^{(1,q)}(z)$ is known explicitly.

We observe that the statistics for the trapping of k walkers is directly related to the kth power of the one-walker numbers $R_n^{(1,q)}$. The simplicity of this result reflects the fact that the walkers under consideration have been supposed to be independent from each other. The convolution relation (25) expresses this independence in a different form.

Finally, we find it convenient to introduce another family of generating functions closely related to the $\{R^{(k,q)}(z)\}$. The kth member of this family, which we denote by $S^{(k,q)}(z)$, generates the kth power of $R_n^{(1,q)} + q$, i.e.,

$$S^{(k,q)}(z) = \sum_{n=0}^{\infty} (R_n^{(1,q)} + q)^k z^n, \qquad k = 0, 1, 2, \ldots \tag{26}$$

Comparing (24) with (26), we note that

$$S^{(k,q)}(z) = \sum_{l=0}^{k} \binom{k}{l} q^{k-l} R^{(l,q)}(z) \tag{27}$$

or, alternatively, that

$$R^{(k,q)}(z) = \sum_{l=0}^{k} (-1)^{k-l} \binom{k}{l} q^{k-l} S^{(l,q)}(z) \tag{28}$$

where

$$R^{(0,q)}(z) \equiv S^{(0,q)}(z) = 1/(1 - z) \tag{29}$$

Combining (27) and (28) with (23), we obtain

$$\langle n\rangle^{(k,q)} = \frac{1}{(N-q)^k}\frac{d}{dz}(1 - z)S^{(k,q)}(z)\bigg|_{z=1} - \sum_{l=1}^{k-1} \binom{k}{l}\left(\frac{q}{N-q}\right)^{k-l} \langle n\rangle^{(l,q)} \tag{30}$$

Also,

$$S^{(k,q)}(z) = \frac{1}{2\pi i} \int_{\Gamma_0} S^{(1,q)}(z'^{-1}) S^{(k-1,q)}(zz') z'^{-1} \, dz' \tag{31}$$

and, in particular,

$$S^{(1,1)}(z) = [(1 - z)^2 P(\mathbf{0}; z)]^{-1} \tag{32a}$$

$$S^{(1,2)}(z) = 2\{(1 - z)^2 [P(\mathbf{0}; z) + P(\mathbf{s}_2 - \mathbf{s}_1; z)]\}^{-1} \tag{32b}$$

The simpler form which $S^{(1,q)}(z)$ has in comparison with $R^{(1,q)}(z)$ makes the convolution relation (31) easier to handle than Eq. (25). For this reason, Eq. (30) is a practical alternative to Eq. (23) in the evaluation of multiple-walker lifetimes.

3. MULTIPLE TRAPPING FOR CONTINUOUS TIME VARIABLE

The preceding results can be used as a basis for the analysis of the multiple-trapping problem for continuous-time random walks. It is within the framework of a continuous-time variable that we can relax assumption (iii) of Section 1 on simultaneous stepping at regular time intervals.

Following Montroll,[11] we shall assume that jumps are made at random times $t_1, t_2, \ldots$, where the random variables

$$T_1 = t_1, \qquad T_2 = t_2 - t_1, \ldots, \qquad T_n = t_n - t_{n-1}$$

have a common density $\psi(t)$. Thus, $\psi(t)$ is the probability density for a step to occur at time t. For convenience we also introduce the probability densities $\{\psi_n(t)\}$ for the occurrence of the nth step at time t. These densities are related to each other by the convolution integral

$$\psi_0(t) = \delta(t), \qquad \psi_n(t) = \int_0^t d\tau \, \psi(t - \tau) \psi_{n-1}(\tau), \qquad n = 1, 2, \ldots \tag{33}$$

As in the previous section, we consider first the problem of one walker and q traps. The probability density $F(\mathbf{s}_0, t)$ that a walker, starting his walk from $\mathbf{s}_0 \notin Q$, is trapped at time t is given by

$$F(\mathbf{s}_0, t) = \sum_{n=1}^{\infty} F_n(\mathbf{s}_0) \psi_n(t) \tag{34}$$

and the Laplace transform of $F(\mathbf{s}_0, t)$,

$$\hat{F}(\mathbf{s}_0, u) = \int_0^{\infty} dt \, e^{-uT} F(\mathbf{s}_0, t) \tag{35}$$

has the form

$$\hat{F}(s_0, u) = \sum_{n=1}^{\infty} F_n(\mathbf{s}_0)[\hat{\psi}(u)]^n = F(\mathbf{s}_0; \hat{\psi}(u)) \tag{36}$$

where $\hat{\psi}(u)$ is the Laplace transform of $\psi(t)$,

$$\hat{\psi}(u) = \int_0^{\infty} dt\, e^{-ut}\psi(t), \qquad \hat{\psi}(0) = 1 \tag{37}$$

and $F(\mathbf{s}_0; \hat{\psi}(u))$ is the generating function (1) with z set equal to $\hat{\psi}(u)$. Assuming again that the walker has the same probability of starting his walk from any nontrapping point, the mean time for trapping is given by

$$\langle t \rangle^{(1,q)} = \frac{1}{N-q} \sum_{\mathbf{s}_0 \notin Q} \int_0^{\infty} dt\, tF(\mathbf{s}_0, t) \tag{38}$$

or, in terms of $\hat{F}(\mathbf{s}_0, u)$, by

$$\langle t \rangle^{(1,q)} = \frac{1}{N-q} \sum_{\mathbf{s}_0 \notin Q} \frac{\partial}{\partial u} \hat{F}(\mathbf{s}_0, u) \Big|_{u=0} \tag{39}$$

Now, we introduce expressions analogous to $R_n^{(1,q)}$ and its generating function $R^{(1,q)}(z)$. These are

$$T^{(1,q)}(t) = \sum_{\mathbf{s}_0 \notin Q} \int_0^t d\tau\, F(\mathbf{s}_0, \tau) \tag{40}$$

and its Laplace transform

$$\hat{T}^{(1,q)}(u) = \int_0^{\infty} dt\, e^{-ut} T^{(1,q)}(t) = \sum_{\mathbf{s}_0 \notin Q} u^{-1} F(\mathbf{s}_0; \hat{\psi}(u)) \tag{41}$$

which, in terms of $R_n^{(1,q)}$ and $R^{(1,q)}(z)$ take the form

$$T^{(1,q)}(t) = \int_0^t d\tau\, \Psi(t-\tau) \sum_{n=0}^{\infty} R_n^{(1,q)} \psi_n(\tau) \tag{42}$$

and

$$\hat{T}^{(1,q)}(u) = \hat{\Psi}(u) R^{(1,q)}(\hat{\psi}(u)) \tag{43}$$

where $\Psi(t)$,

$$\Psi(t) = \int_t^{\infty} d\tau\, \psi(\tau) \tag{44}$$

is the probability that the walker remains fixed in the time interval $(0, t)$ and $\hat{\Psi}(u)$ is its Laplace transform. From Eqs. (39) and (41) we obtain for the mean lifetime $\langle t \rangle^{(1,q)}$ the expression

$$\langle t \rangle^{(1,q)} = -\frac{1}{N-q} \frac{d}{du} u\hat{T}^{(1,q)}(u) \Big|_{u=0} \tag{45}$$

and this together with (43) and (6) leads to the simple (and expected) result[11]

$$\langle t\rangle^{(1,q)} = T\langle n\rangle^{(1,q)} \tag{46}$$

where T is the average time between jumps,

$$T = \int_0^\infty dt\, t\psi(t) \tag{47}$$

The mean lifetime of two walkers on the lattice, with the same density $\psi(t)$, is given by

$$\langle t\rangle^{(2,q)} = \frac{1}{(N-q)^2} \sum_{\mathbf{s}_0,\mathbf{r}_0 \notin Q} \int_0^t dt\, tG(\mathbf{r}_0, \mathbf{s}_0, t) \tag{48}$$

where

$$G(\mathbf{r}_0, \mathbf{s}_0, t) = F(\mathbf{s}_0, t) \int_0^t d\tau\, F(\mathbf{r}_0, \tau) + F(\mathbf{r}_0, t) \int_0^t d\tau\, F(\mathbf{s}_0, \tau) \tag{49}$$

But since

$$\sum_{\mathbf{s}_0,\mathbf{r}_0 \notin Q} G(\mathbf{r}_0, \mathbf{s}_0, t) = (d/dt)[T^{(1,q)}(t)]^2 \tag{50}$$

we also have

$$\langle t\rangle^{(2,q)} = -\frac{1}{(N-q)^2} \frac{d}{du} uT^{(2,q)}(u)\bigg|_{u=0} \tag{51}$$

where

$$\hat{T}^{(2,q)}(u) = \frac{1}{2\pi i} \int_{\gamma - i\infty}^{\gamma + i\infty} \hat{T}^{(1,q)}(u')\hat{T}^{(1,q)}(u-u')\, du' \tag{52}$$

and where γ is such that all the singularities of $\hat{T}^{(1,q)}(u)$ lie to the left of the line $\operatorname{Re} u = \gamma$.

The above results can be easily generalized to the case of k walkers, all with the same density $\psi(t)$. We obtain the following general expression for their mean lifetime:

$$\langle t\rangle^{(k,q)} = -\frac{1}{(N-q)^k} \frac{d}{du} u\hat{T}^{(k,q)}(u)\bigg|_{u=0} \tag{53}$$

where the function $\hat{T}^{(k,q)}(u)$ satisfies the recursion relation

$$\hat{T}^{(k,q)}(u) = \frac{1}{2\pi i} \int_{\gamma - i\infty}^{\gamma + i\infty} \hat{T}^{(1,q)}(u')\hat{T}^{(k-1,q)}(u-u')\, du' \tag{54}$$

These, together with (43) and the results of the previous section, permit the evaluation of $\langle t\rangle^{(k,q)}$ from the basic functions $p(\mathbf{s})$ and $\psi(t)$ that specify the nature of the walks. In particular, the choice $\psi(t) = \delta(t - T)$, which corresponds to simultaneous stepping at regular times, implies that

$$T^{(k,q)}(t) = R_n^{(k,q)}, \qquad nT \leqslant t < (n+1)T \tag{55}$$

or, in Laplace space, that

$$\begin{aligned}\hat{T}^{(k,q)}(u) &= \sum_{n=1}^{\infty} u^{-1}e^{-nTu}(1 - e^{-Tu})R_n^{(k,q)} \\ &= u^{-1}(1 - e^{-Tu}) \sum_{n=1}^{\infty} R_n^{(k,q)} e^{-nTu} \\ &= \hat{\Psi}(u) R^{(k,q)}(\hat{\psi}(u))\end{aligned} \tag{56}$$

which in turn leads to the result

$$\langle t \rangle^{(k,q)} = T\langle n \rangle^{(k,q)} \tag{57}$$

Equation (57) is in general not true for $k > 1$.

An alternative, and more practical, expression for the mean lifetime $\langle t \rangle^{(k,q)}$ is

$$\langle t \rangle^{(k,q)} = -\frac{1}{(N-q)^k} \frac{d}{du} u\hat{U}^{(k,q)}(u)\bigg|_{u=0} - \sum_{l=1}^{k=1} \binom{k}{l} \left(\frac{q}{N-q}\right)^{k-l} \langle t \rangle^{(l,q)} \tag{58}$$

where

$$\hat{U}^{(k,q)} = \frac{1}{2\pi i} \int_{\gamma - i\infty}^{\gamma + i\infty} \hat{U}^{(1,q)}(u')\hat{U}^{(k-1,q)}(u - u')\, du' \tag{59}$$

and where

$$\hat{U}^{(1,q)}(u) = \hat{\Psi}(u) S^{(1,q)}(\hat{\psi}(u)) \tag{60}$$

4. MEAN ABSORPTION TIME FOR k WALKERS ON ONE-DIMENSIONAL RINGS

We are now in a position to apply the formalism to a concrete situation, specifically to the example of symmetric walks on a one-dimensional ring with nearest neighbor transitions. In this case one has[11]

$$P(l; z) = \frac{1}{N} \sum_{k=0}^{N-1} \frac{\exp(2\pi i k/N)}{1 - z\cos(2\pi k/N)} = \frac{1}{(1 - z^2)^{1/2}} \frac{W^l + W^{N-l}}{1 - W^N} \tag{61}$$

where

$$W = (1/z)[1 - (1 - z^2)^{1/2}] \tag{62}$$

4.1. Simultaneous Stepping at Regular Time Intervals

We consider first one trap only, and then we extend the results to the case of q equally spaced traps.

One Walker. Montroll[5] has shown that for a ring of N particles with one trap

$$\langle n\rangle^{(1,1)} = N(N+1)/6 \tag{63}$$

Two Walkers. As shown in the appendix, the generating function $P(0; z)$ can be factorized as follows:

$$P(0; z) = \frac{\prod_{j=1}^{M}\{1 - z\cos[2\pi(2j-1)/2N]\}}{\prod_{j=0}^{M}\{1 - z\cos(2\pi j/N)\}} \tag{64}$$

where

$$M = \begin{cases} N/2, & N \text{ even} \\ (N-1)/2, & N \text{ odd} \end{cases} \tag{65}$$

thus allowing Eq. (31), for $k = 2$ and $q = 1$, to be written as

$$\begin{aligned} S^{(2,1)}(z) = \frac{1}{2\pi i}\int_{\Gamma_0} \Bigg\{ & \left[\prod_{j=1}^{M}\left(z' - \cos\frac{2\pi j}{N}\right)\prod_{j=1}^{M}\left(1 - zz'\cos\frac{2\pi j}{N}\right) dz'\right] \\ & \times \left[(z'-1)(1-zz')\prod_{j=1}^{M}\left(z' - \cos\frac{2\pi(2j-1)}{2N}\right)\right. \\ & \times \left.\prod_{j=1}^{M}\left(1 - zz'\cos\frac{2\pi(2j-1)}{2N}\right)\right]^{-1}\Bigg\} \end{aligned} \tag{66}$$

When $|z| < 1$, the singularities of the integrand in (66) lying within the unit circle are simple poles located at $z' = 1$ and at

$$z_l = \cos[2\pi(2l-1)/2N], \qquad l = 1,\ldots, M \tag{67}$$

Therefore, by using the method of residues, we find

$$\begin{aligned} S^{(2,1)}(z) = {} & \frac{\prod_{j=1}^{M}[1 - \cos(2\pi j/N)]}{\prod_{j=1}^{M}\{1 - \cos[2\pi(2j-1)/2N]\}} S^{(1,1)}(z) \\ & + \sum_{l=1}^{M}\frac{1}{(1-z_l)^2}\frac{\prod_{j=0}^{M}[z_l - \cos(2\pi j/N)]}{\prod_{j=1;j\neq l}^{M}\{z_l - \cos[2\pi(2j-1)/2N]\}} \\ & \times S^{(1,1)}(z_l z), \qquad |z| < 1 \end{aligned} \tag{68}$$

Now, from Eqs. (32a) and (64)

$$\begin{aligned} \frac{\prod_{j=1}^{M}[1 - \cos(2\pi j/N)]}{\prod_{j=1}^{M}\{1 - \cos[2\pi(2j-1)/2N]\}} &= \lim_{z\to 1}[(1-z)P(0; z)]^{-1} \\ &= 1 + \lim_{z\to 1}\sum_{n=1}^{\infty}(R_n^{(1,1)} - R_{n-1}^{(1,1)})z^n = N \end{aligned} \tag{69}$$

where the last equality follows from the requirement that, $R_n^{(1,1)}$ being the average number of distinct sites visited in an n-step walk, $\lim_{n\to\infty} R_n^{(1,1)} = N - 1$. In the appendix it is also shown that the prefactor of $S^{(1,1)}(z_l z)$ can be reduced to the form

$$\frac{1}{(1 - z_l)^2} \frac{\prod_{j=0}^{M} [z_l - \cos(2\pi j/N)]}{\prod_{j=1;j\neq l}^{M} \{z_l - \cos[2\pi(2j - 1)/2N]\}} = -\frac{2}{N}\frac{2 + z_l}{1 - z_l} \tag{70}$$

Thus,

$$S^{(2,1)}(z) = NS^{(1,1)}(z) - \frac{2}{N}\sum_{l=1}^{N} \frac{1 + z_l}{1 - z_l} S^{(1,1)}(z_l z), \qquad |z| < 1 \tag{71}$$

Finally, by combining Eq. (30) for $k = 2$ and $q = 1$ with Eq. (71), and taking into account that $dS^{(1,1)}(z)/dz$ is analytic for $|z| < 1$, we obtain for the lifetime $\langle n\rangle^{(2,1)}$ the expression

$$\langle n\rangle^{(2,1)} = \frac{N - 2}{N - 1}\langle n\rangle^{(1,1)} + \frac{2}{N(N - 1)^2}\sum_{l=1}^{M} \frac{1 + z_l}{1 - z_l} S^{(1,1)}(z_l) \tag{72}$$

k **Walkers.** By substituting Eq. (71) into (31), for $k = 3$ and $q = 1$ one can readily see that, when $|z| < 1$, all the singularities contributing to the resulting contour integral come again from the function $S^{(1,1)}(z'^{-1})$, i.e., they are simple poles located at $z' = 1$ and at the z_l defined by (32). By iteration, we observe that this situation is not altered for larger values of k, and, hence, we conclude that

$$S^{(k,1)}(z) = NS^{(k-1,1)}(z) - \frac{2}{N}\sum_{l=1}^{M} \frac{1 + z_l}{1 - z_l} S^{(k-1,1)}(z_l z), \qquad k = 2, 3, \dots, \quad |z| < 1 \tag{73}$$

Furthermore, repeated substitution of Eq. (73) in itself yields

$$S^{(k,1)}(z) = \sum_{j=0}^{k=1} (-1)^j 2^j \binom{k - 1}{j} N^{k-(2j+1)} \sum_{l_1=\cdots=l_j=1}^{M} \times \prod_{m=0}^{j} [(1 + z_{l_m})/(1 - z_{l_m})] S^{(1,1)}(z_{l_1} \cdots z_{l_j} z) \tag{74}$$

where $z_{l_0} \equiv 0$ and

$$z_{l_m} = \cos[2\pi(2l_m - 1)/2N], \qquad l_m = 1, \dots, M \tag{75}$$

Therefore

$$\frac{1}{(N - 1)^k}\frac{d}{dz}(1 - z)S^{(k,1)}(z)\bigg|_{z=1} = \left(\frac{N}{N - 1}\right)^{k-1}\langle n\rangle^{(1,1)} + \left(\frac{N}{N - 1}\right)^k \sum_{j=1}^{k-1} (-1)^{j+1} 2^j \binom{k - 1}{j} N^{-(2j+1)} T_j \tag{76}$$

where

$$T_j = \sum_{l_1=\cdots=l_j=1}^{M} \prod_{m=1}^{j} [(1+z_{l_m})/(1-z_{l_m})] S^{(1,1)}(z_{l_1}\cdots z_{l_j}) \tag{77}$$

The mean lifetime for the k walkers is obtained by combining Eqs. (30) [for $q = 1$] and (76).

Finally, we derive an expression for $\langle n\rangle^{(k,1)}$ which is valid for large N and small k. The largest contribution to T_j, above, comes from the term $l_m = 1$, $m = 1,\ldots,j$. For this, when N is large, and with the help of the expression $z_1 \sim 1 - \pi^2/2N^2 + \cdots$, we find

$$t_j = \left(\frac{1+z_1}{1-z_1}\right)^j S^{(1,1)}(z_1{}^j) \sim \frac{2^{2(j+1)}N^{2j+3}}{j^{3/2}\pi^{2j+3}} \frac{1-\exp(-j^{1/2}\pi)}{1+\exp(-j^{1/2}\pi)} \tag{78}$$

Also, for the second largest contributions to Eq. (77), we have

$$\begin{aligned} u_j &= j\left(\frac{1+z_1}{1-z_1}\right)^{j-1} \frac{1+z_2}{1-z_2} S^{(1,1)}(z_1^{j-1}z_2) \\ &\sim \frac{2^{2(j+1)}jN^{2j+3}}{3^2(3^2+j-1)^{3/2}\pi^{2j+3}} \frac{1-\exp[(3^2+j-1)^{1/2}\pi]}{1+\exp[(3^2+j-1)^{1/2}\pi]} \end{aligned} \tag{79}$$

Similar expressions can be obtained for

$$v_j = j\left(\frac{1+z_1}{1-z_1}\right)^{j-1} \frac{1+z_3}{1-z_3} S^{(1,1)}(z_1^{j-1}z_3) \tag{80}$$

and

$$w_j = j(j-1)\left(\frac{1+z_1}{1-z_1}\right)^{j-2} \left(\frac{1+z_2}{1-z_2}\right)^2 S^{(1,1)}(z_1^{j-2}z_2{}^2) \tag{81}$$

Relative values taken by these quantities for $1 \leqslant j \leqslant 8$ are shown in Table I. We can see from this table that, when we consider the trapping of a small

Table I. Relative Values of the First Few Terms in Eq. (77) as Defined by Eqs. (78)–(81)

j	u_j/t_j	v_j/t_j	w_j/t_j
1	0.0044	0.0003	—
2	0.0203	0.0017	0.0009
3	0.0478	0.0045	0.0046
4	0.0858	0.0086	0.0133
5	0.1327	0.0143	0.0287
6	0.1872	0.0215	0.0528
7	0.2480	0.0300	0.0871
8	0.3143	0.0400	0.1330

Table II. Values for the Coefficient a_k for the Mean Lifetime of k Walkers[a] Calculated from Eq. (83)

k	1	2	3	4	5	6
a_k	0.1666	0.2630	0.3294	0.3798	0.4202	0.4534

[a] $\langle n\rangle^{(k,1)} \sim a_k N^2$.

number of walkers on a large ring, the quantities designated by t_j and u_j will account for nearly all of T_j (with k no greater than six, t_j and u_j amount to over 96% of T_j). And, since the second term in Eq. (30), when $q = 1$, is necessarily of lower order in N than the first term [Eq. (76)], we have for the mean lifetime $\langle n\rangle^{(k,1)}$ the result

$$\langle n\rangle^{(k,1)} \sim a_k N^2, \qquad \text{large } N \tag{82}$$

where

$$a_k = \tfrac{1}{6} + \sum_{j=1}^{k-1} (-1)^{j+1} 2^{3j+2} \pi^{-(2j+3)} \binom{k-1}{j} \times \left[j^{-3/2} \frac{1 - \exp(-j^{1/2}\pi)}{1 + \exp(-j^{1/2}\pi)} + 3^{-2} j (3^2 + j - 1)^{-3/2} \frac{1 - \exp[-(3^2 + j - 1)^{1/2}\pi]}{1 + \exp[-(3^2 + j - 1)^{1/2}\pi]} + \cdots \right] \tag{83}$$

In Table II we give values of a_k for $k = 1,\ldots, 6$. From these we observe that the difference in lifetimes $\langle n\rangle^{(k,1)} - \langle n\rangle^{(k-1,1)}$ decreases as k increases. This, of course, is due to the fact that, on average, when a walker is trapped the remaining walkers have already made a number of steps.

***k* Walkers with *q* Equally Spaced Traps.** The analytical discussion of the case of q arbitrarily located traps leads to more complicated expressions than those for one trap. However, when the traps are spaced regularly, with N/q integer, a simple relation follows immediately between $\langle n\rangle^{(k,q)}$ and $\langle n\rangle^{(k,1)}$. In this case, the N-site ring consists of q intervals separated by traps, each of which contains $(N/q) - 1$ nontrapping points. Each of the k walkers has the same probability of starting at any interval, but once he starts on a given interval, he cannot escape from it, and thus his trapping is equivalent to that of a walker on a ring of N/q points with only one trap. Since the walkers are independent, we conclude that the mean lifetime of k walkers on an N-site ring with q equally spaced traps is the same as that of k walkers on a ring with N/q points, one of which is a trap, i.e.,

$$\langle n\rangle^{(k,q)}_{N\ \text{sites}} = \langle n\rangle^{(k,1)}_{N/q\ \text{sites}} \tag{84}$$

The same result can be derived analytically from our general formulas. For $Q = \{0, N/q, \ldots, (q-1)N/q\}$, Eqs. (9), (12), and (13), together with (16) and (61), yield

$$R_N^{(1,q)}(z) = \frac{q}{1-z}\left\{\left[(1-z)\sum_{j=0}^{q-1} P_N\left(\frac{jN}{q}; z\right)\right]^{-1} - 1\right\}$$

$$= \frac{q}{1-z}\{[(1-z)P_{N/q}(0; z)]^{-1} - 1\} = qR_{N/q}^{(1,1)}(z) \tag{85}$$

and from this and Eqs. (25) and (23) we obtain Eq. (84).

It is interesting to note that the validity of (84) is not restricted to either one-dimensional walks or nearest neighbor transitions. As long as the walkers are independent and the traps are regularly spaced, the q-trap problem can always be shown to be equivalent to a one-trap problem on a lattice with N/q sites (and with periodic boundary conditions).

4.2. Exponentially Distributed Stepping Times

We examine now the trapping of k walkers on a ring containing one trap when all of the walkers step according to the exponential density

$$\psi(t) = (1/T)e^{-t/T} \tag{86}$$

From (86) we have

$$\hat{\psi}(u) = 1/(1 + uT) \tag{87a}$$

$$\hat{\Psi}(u) = T/(1 + uT) \tag{87b}$$

and thus (60) becomes

$$\hat{U}^{(1,1)}(u) = \frac{T}{1+uT} S^{(1,1)}\left(\frac{1}{1+uT}\right) \tag{88}$$

Due to the above relationship between $\hat{U}^{(1,1)}$ and $S^{(1,1)}$, the evaluation of $\hat{U}^{(k,1)}$ by means of the recursion formula (59) is very similar to that already presented for $S^{(k,1)}$. Thus, from Eqs. (88) and (32a) and the factorization (64), we have

$$\hat{U}^{(2,1)}(u) = \frac{1}{2\pi i}\int_{-i\infty}^{i\infty} \frac{\prod_{j=1}^{M}[1 + u'T - \cos(2\pi j/N)]}{u'\prod_{j=1}^{M}\{1 + u'T - \cos[2\pi(2j-1)/2N]\}} \times \hat{U}^{(1,1)}(u - u')\, du' \tag{89}$$

When $\operatorname{Re} u > 0$, the singularities of the integrand in (89) lying to the left of the line $\operatorname{Re} u' = 0$ are simple poles located at $u' = 0$ and at

$$u_l = -T^{-1}\{1 - \cos[2\pi(2l-1)/2N]\}, \qquad l = 1, \ldots, M \tag{90}$$

and therefore,

$$\hat{U}^{(2,1)}(u) = \frac{\prod_{j=1}^{M}[1 - \cos(2\pi j/N)]}{\prod_{j=1}^{M}\{1 - \cos[2\pi(2j-1)/2N]\}}\hat{U}^{(1,1)}(u) + \sum_{l=1}^{M}\frac{\prod_{j=1}^{M}[1 + Tu_l - \cos(2\pi j/N)]}{Tu_l\prod_{j=1;j\neq l}^{M}\{1 + Tu_l - \cos[2\pi(2j-1)/2N]\}} \times \hat{U}^{(1,1)}(u - u_l), \qquad \text{Re}\, u > 0 \tag{91}$$

Since

$$1 + Tu_l = \cos[2\pi(2l-1)/2N] = z_l \tag{92}$$

by means of Eqs. (69) and (70), Eq. (91) is reduced to

$$\hat{U}^{(2,1)}(u) = N\hat{U}^{(1,1)}(u) + \frac{2}{N}\sum_{l=1}^{M}\frac{2 + Tu_l}{Tu_l}\hat{U}^{(1,1)}(u - u_l), \qquad \text{Re}\, u > 0 \tag{93}$$

And, by following an argument parallel to the one that led to Eqs. (73) and (74), we find

$$\hat{U}^{(k,1)}(u) = N\hat{U}^{(k,1)}(u) + \frac{2}{N}\sum_{l=1}^{M}\frac{2 + Tu_l}{Tu_l}\hat{U}^{(k-1,1)}(u - u_l) = \sum_{j=0}^{k-1}2^j\binom{k-1}{j}N^{k-(2j+1)}\sum_{l_1=\cdots=l_j=1}^{M} \times \prod_{m=0}^{j}\frac{2 + Tu_{l_m}}{Tu_{l_m}}\hat{U}^{(1,1)}(u - u_{l_1} - \cdots - u_{l_j}), \qquad \text{Re}\, u > 0 \tag{94}$$

Also,

$$-\frac{1}{(n-1)^k}\frac{d}{du}u\hat{U}^{(k,1)}(u)\bigg|_{u=0} = \left(\frac{N}{N-1}\right)^{k-1}\langle t\rangle^{(1,1)} + \left(\frac{N}{N-1}\right)^{k}\sum_{j=1}^{k-1}2^j\binom{k-1}{j}N^{-(2j+1)}A_j \tag{95}$$

where

$$A_j = \sum_{l_1=\cdots=l_j=1}^{M}\prod_{m=1}^{j}\frac{2 + Tu_{l_m}}{Tu_{l_m}}\hat{U}^{(1,1)}(-u_{l_1} - \cdots - u_{l_j}) = (-1)^{j+1}T\sum_{l_1=\cdots=l_j=1}^{M}\prod_{m=1}^{j}\frac{1 + z_{l_m}}{1 - z_{l_m}} \times \frac{1}{j + 1 - z_1 - \cdots - z_j}S^{(1,1)}\left(\frac{1}{j + 1 - z_1 - \cdots - z_j}\right) \tag{96}$$

Table III. Values for the Mean Lifetime $\langle n\rangle^{(2,1)}$ for Regular Stepping[a] and for the Difference in Lifetimes $T^{-1}[\langle t\rangle^{(2,1)} - T\langle n\rangle^{(2,1)}]$ Between Exponential and Regular Stepping Time Laws[b]

N	$\langle n\rangle^{(2,1)}$	$T^{-1}[\langle t\rangle^{(2,1)} - T\langle n\rangle^{(2,1)}]$
4	4.74	0.2870
8	18.18	0.2321
12	40.00	0.2161
16	70.24	0.2084
20	108.89	0.2038
24	155.96	0.2008
40	428.40	0.1947

[a] From Eqs. (30) and (76).
[b] From Eq. (98).

If we now examine the behavior of $\langle t\rangle^{(k,1)}$ for large N, as we did above for $\langle n\rangle^{(k,1)}$, we find that

$$\langle t\rangle^{(k,1)} \sim T\langle n\rangle^{(k,1)}, \qquad \text{large } N \tag{97}$$

with $\langle n\rangle^{(k,1)}$ given by Eqs. (82) and (83). This is not surprising, for, when N is large, most walkers start their walk far from the trap, and then they must step a large number of times before trapping. These walks, on average, are well represented by simultaneous stepping at regular time intervals of magnitude T, the average time between jumps. This interpretation of the limiting form (97) is not restricted to the exponential density we considered, and hence, we expect it to be of more general validity.

In Table III we give values, for various sizes of lattices, of the difference in lifetimes $\langle t\rangle^{(2,1)} - T\langle n\rangle^{(2,1)}$, i.e.,

$$\langle t\rangle^{(2,1)} - T\langle n\rangle^{(2,1)} = \frac{2T}{N(N-1)^2}\sum_{l=1}^{M}\frac{1+z_l}{1-z_l}\left[\frac{1}{2-z_l}S^{(1,1)}\left(\frac{1}{2-z_l}\right) - S^{(1,1)}(z_l)\right] \tag{98}$$

5. DISCUSSION

Under the basic assumption of walker independence, we have shown that the mean absorption time of a set of walkers is determined by the statistics of only one walker. This result is conveniently expressed in terms of a family of generating functions related by a recursion formula, and by means of which one constructs k-walker lifetimes from one-walker properties.

When one considers the case of only one trap, the kth member of this family generates the kth power of the average number of distinct sites visited after time t.

In particular, for one-dimensional walks, we found that, although there are no order-of-magnitude changes, augmenting the number of walkers on the lattice has a significant effect on $\langle n\rangle^{(k,1)}$ when k is small. We also found that the difference in lifetimes $\langle t\rangle^{(k,1)} - T\langle n\rangle^{(k,1)}$ between exponentially distributed stepping times and simultaneous stepping at regular intervals disappears as N, the number of lattice sites, increases. This limiting property should hold for arbitrary $\psi(t)$, as well as for lattices containing more than one trap, provided they are widely separated. Similar properties are to be expected in higher dimensional lattices. However, to prove this, one would have to work a little harder in establishing formulas similar to Eq. (76) or (95), since the generating function $P(\mathbf{s}; z)$ for $d > 1$ is not easily expressed in a closed form.

It would be of interest to extend the multiple trapping problem studied here to the case of interacting walkers. Some effects of exciton motion on molecular arrays, such as deexcitation of the network through fluorescence produced by exciton fusion, could be modeled in terms of interacting walkers.

APPENDIX. FACTORIZATION OF $P(0; z)$ AND DERIVATION OF EQ. (70)

The identity[12]

$$\prod_{j=1}^{N-1}\left[x^2 - 2xy\cos\left(\alpha + \frac{2\pi j}{N}\right) + y^2\right] = x^{2N} - 2x^N y^N \cos\alpha N + y^{2N} \tag{A1}$$

when N is even, and with the choice $x^2 + y^2 = 1$, $2xy = z$, and $\alpha = -\pi/N$, is reduced to

$$\prod_{j=1}^{N/2}\{1 - z\cos[2\pi(2j-1)/2N]\} = x^N + y^N \tag{A2}$$

If instead we choose $\alpha = 0$, we have

$$\prod_{j=0}^{N/2}[1 - z\cos(2\pi j/N)] = (1 - z^2)^{1/2}(x^N - y^N) \tag{A3}$$

Now, since

$$y/x = [1 - (1 - z^2)^{1/2}]/z \tag{A4}$$

dividing (A2) by (A3) yields the factorization (64).

On the other hand, if we choose $x^2 + y^2 = z$, $2xy = 1$, with first $\alpha = -\pi/N$ and then $\alpha = 0$, the identity (A1) becomes, respectively,

$$\prod_{j=1}^{N/2} \{z - \cos[2\pi(2j - 1)/2N]\} = x^N + y^N \tag{A5}$$

and

$$\prod_{j=0}^{N/2} [z - \cos(2\pi j/N)] = (z^2 - 1)^{1/2}(x^N - y^N) \tag{A6}$$

with

$$x^2 = \tfrac{1}{2}[z + (z^2 - 1)^{1/2}] \tag{A7a}$$

and

$$y^2 = \tfrac{1}{2}[z - (z^2 - 1)^{1/2}] \tag{A7b}$$

Substitution of (A5)–(A7) into

$$\frac{\prod_{j=0}^{N/2} [z_l - \cos(2\pi j/N)]}{\prod_{j=1;j\neq l}^{N/2} \{z_l - \cos[2\pi(2j - 1)/2N]\}} = \frac{\prod_{j=0}^{N/2} [z_l - \cos(2\pi j/N)]}{[(\partial/\partial z) \prod_{j=1}^{N/2} \{z - \cos[2\pi(2j - 1)/2N]\}]_{z=z_l}} \tag{A8}$$

leads to (70).

Similarly with N odd.

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