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Random walks and generalized master equations with internal degrees of freedom

(stochastic processes/transport phenomena)

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ABSTRACT **We present an extension of the continuous-time random walk formalism to include internal states and to establish the connection to generalized master equations with internal states. The theory allows us to calculate physical observables from which we can extract the characteristic parameters of the internal states of the system under study.**

Physical systems can be described at different levels. In the simplest models the system is considered to be a structureless whole. Attempts to understand nature on finer scales incorporate sharper details of internal structure. The definition and identification of the structural elements are naturally dictated by the system under investigation. Consequently, a maximal degree of detail (microscopicity) usually exists, governed by characteristic parameters of the subject phenomenon (e.g., range of forces, correlation lengths, relaxation times, etc.).

Our aim is to describe, through examples, a manner of decomposing systems into internal states and to present a rich random walk formalism capable of characterizing internal states and unusual relaxation effects. The total scheme may be viewed as an effective scaling in terms of which fundamental processes may be renormalized and their dynamics analyzed.

For our first example, we note that atomic clusters adsorbed on crystalline surfaces may exist in several geometric configurations (Fig. 1, upper panels). During configurational transitions, cluster centroids experience a lattice random walk (1) displaying several internal states per unit cell (details to be published elsewhere) (Fig. 1, lower panels).

Random-walk lattice sites need not represent spatial displacements of the system variables but may merely model the state of a counter, such as the flux of tracer ions through a neural membrane. The positions of tracer ions within a channel represent the internal states of the channel, and the counter changes when a tracer ion leaves the membrane (2).

Other types of membrane transport are enzyme-assisted as in some cell wall transport (3). The ligand-membrane (enzyme) complex may exist in several distinct states as intermediates in the transport. A single ligand being transported through the membrane becomes equivalent to a step to the right in a one-dimensional walk. If two different ligand species may penetrate the membrane, the transport process maps onto a two-dimen- displaying several internal states per unit cell (details to be published elsewhere) (Fig. 1, lower panels).

Finally, transient photocurrents in certain amorphous materials used as xerographic films can be modeled as the hopping of localized charges over a random distribution of spatial sites or random potential depths (4). In this last case, a periodic lattice is mathematically imposed, such that the number of sites per unit cell is large. The motions in and out of a cell are treated as random events. So many possible states exist for a carrier inside a cell that it is almost hopeless to try to identify them. Therefore, the germane characteristic of the process is specified by a single waiting time distribution function for the leaving of structureless unit cells. This description is similar to proceeding from a microscopic to a macroscopic description, such as in the hydrodynamic description of a fluid.

A unifying feature of the above models is a continuous-time random walk on a lattice. An appropriate mathematical technique for the investigation of such processes was described a number of years ago (5) and has been further developed and applied more recently by several researchers (6–10). However, internal states were not included in the above analyses. Lattice random walks in which the walker can exist in several possible spatial states per unit cell, but in which steps are taken in regular time intervals, have been analyzed in an application to photosynthesis modeling (11). Weiss (12) recently discussed several aspects of random walks by a walker capable of existing in either of two states.

In this paper, we extend the continuous-time random walk formalism to include internal states and to establish a connection to generalized master equations. The physical observables of the above-mentioned phenomena are typically mean distances, fluxes, and concentrations and their fluctuations. The theory is amenable to these calculations. Moreover, the theory offers methods of analysis from which information about the transition rates between internal states can be found from the experimental data.

DESCRIPTION OF MODEL

Let us consider a d-dimensional space lattice of individual cells, each being identified by a vector

$$s = (s_1, s_2, \ldots, s_d) \quad \text{with} \quad s_\alpha = 1,2,\ldots,N_\alpha. \qquad [1]$$

Generally we will be concerned with periodic boundary conditions

$$(s_1 + N_1, s_2 + N_2, \ldots, s_d + N_d) \equiv (s_1, s_2, \ldots, s_d). \qquad [2]$$

Many applications involve the limiting case of infinite lattices with $N_\alpha \to \infty$ for all α or perhaps for all α but one. The states available to the walker will be chosen to be identical in each cell and will be identified by an index j with $j = 1, 2, \ldots . m$.

The basic quantity of concern is the probability,

$$P_{j,j_0}(s, s_0; t), \qquad [3]$$

that a walker originally created at $t = 0$ in lattice cell s_0 in state j_0 will be in cell s in state j at time t. The transition $(j_0, s_0) \to (j, s)$ is achieved after a number of individual steps by the walker, each being characterized by the function

$$\psi_{j,j'}(s, s'; \tau) \equiv F_{j,j'}(s, s'; \tau)\psi_{j'}(\tau) \qquad [4a]$$

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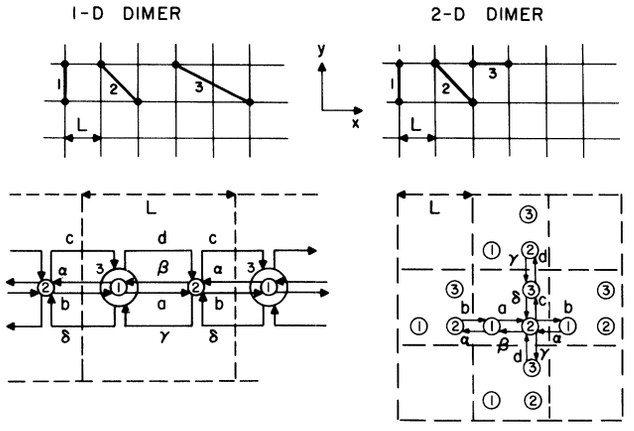

FIG. 1. *Left,* One-dimensional dimer migration. *Upper,* Three possible spatial configurations of a dimer (filled circles connected by heavy line) moving along the x direction (the allowed equivalent mirror image configurations are not included in the figure). If only states 1 and 2 are allowed, this is a two-state dimer; if all states are allowed, it is a three-state dimer. *Lower,* The random-walk lattice describing the motion of the dimer centroid in upper panel. The unit cell is denoted by broken lines and the states by circles, numbered, respectively. Lettered arrows indicate transitions to and from states. Note that transition rates connecting states can be different for transitions to the left or right (i.e., $a \neq \alpha$, etc.). Also, the centroid location is the same for states 1 and 3; however, they are distinguished by different transition rates.

Right. Two-dimensional dimer migration. *Upper,* Spatial configurations. *Lower,* Random-walk lattice.

in which $\psi_j(\tau)$ is the waiting time density function between transitions when in state j (a time which we postulate to be independent of the lattice points s). The F function is the probability of $(j',s') \rightarrow (j,s)$ in one step at time τ when a step is taken. We have the normalization

$$\sum_{j,s} F_{jj'}(s,s';\tau) = 1 \quad \text{and} \quad \int_0^\infty \psi_j(\tau)d\tau = 1. \qquad [4b]$$

Thus, Eq. **4a** is the probability per unit time that, upon arrival at s' in state j', a walker will, after time τ, have just taken a step to s, arriving at that cell in state j. We shall finally be able to express the required elements of the matrix

$$P(s,s_0;t) \equiv \begin{pmatrix} P_{11}(s_1,s_0;t) \ldots P_{1m}(s,s_0;t) \\ \cdots\cdots \\ P_{m1}(s,s_0;t) \ldots P_{mm}(s,s_0;t) \end{pmatrix} \qquad [5]$$

in terms of those of the matrix

$$\psi(s,s';\tau) \equiv \begin{pmatrix} \psi_{11}(s,s';\tau) \ldots \psi_{1m}(s,s';\tau) \\ \cdots\cdots \\ \psi_{m1}(s,s';\tau) \ldots \psi_{mm}(s,s';\tau) \end{pmatrix}. \qquad [6]$$

Furthermore, we postulate that all cells on our basic lattice are equivalent so that the transition probability associated with any displacement is independent of initial position and depends only upon the displacement vector itself. For example

$$\psi(s,s';\tau) \equiv \psi(s - s';\tau) \qquad [7]$$

$$P(s,s_0;t) \equiv P(s - s_0;t) \text{ etc.} \qquad [8]$$

Our lattice walk is executed through an alternating sequence of steps and pauses, both specified by elements of the matrix $\psi(s - s';\tau)$. Consider the first two steps of the walk, which we assume to take a walker from lattice point s' in state j' to lattice point s where it will arrive in state j in the time interval between τ and $\tau + d\tau$. The probability of the occurrence of this event is

$$Q_{jj'}^{(2)}(s - s';\tau)\,d\tau = \sum_{s'',j''} d\tau \int_0^\tau \psi_{jj''}(s - s'';\tau - \tau'')\psi_{j''j'}(s'' - s';\tau'')d\tau'',$$

which is the weighted average summation over all possible two-step paths of the walker. We also set

$$\psi_{jj}(0,\tau) \equiv 0 \quad \text{if} \quad \tau > 0 \qquad [9]$$

to ensure that the act of no transition is not considered to be a step. The matrix $\psi(s,\tau)$ will always be chosen so that

$$\psi(s,\tau) \rightarrow 0 \quad \text{as} \quad \tau \rightarrow 0 \quad \text{for all} \quad s. \qquad [10]$$

The defining equation for $Q_{jj'}^{(2)}(s - s';\tau)$ is more conveniently written in matrix form with the defined quantities being considered as elements of a matrix:

$$Q^{(2)}(s - s';\tau) = \sum_{s''} \int_0^\tau \psi(s - s'';\tau - \tau'')\psi(s'' - s';\tau'')d\tau''.$$

The n step walk carrying the walker from (j',s') to (j,s) with the precise arrival time τ is then characterized by a matrix which satisfies the recurrence relationship

$$Q^{(n)}(s - s';\tau) = \sum_{s''} \int_0^\tau Q^{(n-1)}(s - s'';\tau - \tau'')\psi(s'' - s';\tau'')d\tau''. \qquad [11]$$

with

$$Q^{(1)}(s,\tau) \equiv \psi(s,\tau). \qquad [12]$$

The initial condition we choose for our problem is

$$P(s,t) \rightarrow \delta_{s,0}\,\delta(t - 0^+)I \quad \text{as} \quad t \rightarrow 0, \qquad [13]$$

I being the identity matrix. We also define a matrix $Q^{(0)}(s,\tau)$ by

$$Q^{(0)}(s,\tau) = \delta_{s,0}\,\delta(t - 0^+)I. \qquad [14]$$

Then the matrix

$$Q(s - s';\tau) = \sum_{n=0}^{\infty} Q^{(n)}(s - s';\tau) \qquad [15]$$

satisfies the same initial condition as $P(s - s';\tau)$. Its matrix elements

$$Q_{jj'}(s - s';\tau) = \sum_{n=0}^{\infty} Q_{jj'}^{(n)}(s - s';\tau) \qquad [16]$$

represent the probability density of a walker starting at (j'',s') and just arriving at (j,s) at time τ, since the required probability is the sum of the probabilities of the independent ways of achieving that arrival— i.e., after no steps, after one step, after two steps, etc.

The matrix $Q(s,\tau)$ is a special value of the generating function

$$Q(s,\tau;z) = \sum_{n=0}^{\infty} z^n Q^{(n)}(s,\tau) \qquad [17]$$

obtained by setting $z = 1$.

We now introduce the finite Fourier transform (in the discrete lattice space $\{s\}$) and the Laplace transform in continuous variable τ for the function of Eq. **11** so that the convolution

theorem can be applied. For a function $F(s)$ we define (with $k_j \equiv 2\pi r_j/N$)

$$f(k) \equiv f(2\pi r_1/N_1,\ldots,2\pi r_d/N_d) \qquad [18a]$$
$$\equiv \frac{1}{N_1 \ldots N_d} \sum_{s_1=1}^{N_1} \cdots \sum_{s_d=1}^{N_d} F(s_1 \ldots s_d) \times \exp\, 2\pi i\left[\frac{s_1 r_1}{N_1} + \cdots + \frac{s_d r_d}{N_d}\right].$$

Also, for a function $G(t)$ we define the Laplace transform to be

$$\tilde{g}(u) \equiv \int_0^\infty e^{-tu} G(t)dt. \qquad [18b]$$

Application of the Fourier and Laplace convolution theorems to the recurrence formula, Eq. **11**, yields

$$\tilde{q}^{(n)}(k,u) = \tilde{q}^{(n-1)}(k,u)\tilde{\psi}(k,u) \qquad [19a]$$

$$\tilde{q}^{(1)}(k,u) = \tilde{\psi}(k,u),\ \tilde{q}^{(0)}(k,u) = I. \qquad [19b]$$

Then

$$\tilde{q}^{(n)}(k,u) = \{\tilde{\psi}(k,u)\}^n I \quad \text{for } n = 0, 1, 2, \ldots \qquad [20]$$

so that, from Eq. **17**,

$$\tilde{q}(k,u;z) = I + z\tilde{\psi} + z^2\tilde{\psi}^2 + \ldots = \{I - z\tilde{\psi}(k,u)\}^{-1}. \qquad [21]$$

The required matrix $P(s,t)$ whose elements were defined by Eq. **3** is immediately available from Eq. **21**. The quantity $P_{jj_0}(s,s_0;t)$ is related to $Q_{jj'}(s - s';\tau)$ through the aid of a function $\Gamma_j(\tau)$ which we define to be the probability that a walker arriving at state (j,s) at time 0 has not changed state in the time interval τ after arrival. Then

$$P_{jj'}(s;t) = \int_0^t \Gamma_j(t - \tau)Q_{jj'}(s,\tau)d\tau. \qquad [22]$$

Since $\psi_j(\tau)$ was defined below Eq. **4a** to be the probability density that a walker will leave state j (from any cell that it might occupy) in time τ

$$\Gamma_j(t) = 1 - \int_0^t \psi_j(t - \tau)d\tau. \qquad [23]$$

The Laplace transform of this quantity is

$$\tilde{\gamma}_j(u) = u^{-1}[1 - \tilde{\psi}_j(u)]. \qquad [24]$$

Then, by taking the Laplace and finite Fourier transform of Eq. **22**, the convolution theorem, and Eq. **21**,

$$\tilde{p}(k,u) = u^{-1}[I - \tilde{\psi}_d(u)]\{I - \tilde{\psi}(k,u)\}^{-1} \qquad [25]$$

in which $\tilde{\psi}_d(u)$ is the diagonal matrix

$$\tilde{\psi}_d(u) \equiv \begin{pmatrix} \tilde{\psi}_1(u) & 0 & \ldots & 0 \\ 0 & \tilde{\psi}_2(u) & \ldots & 0 \\ \ldots & \ldots & \ldots & \ldots \\ 0 & \ldots & \ldots & \tilde{\psi}_n(u) \end{pmatrix} \qquad [26]$$

Our initially stated problem is solved by constructing the inverse Laplace, finite Fourier transform of Eq. **25** (with $k_j \equiv 2\pi r_j/N_j$); $\mathcal{L}^{-1}$ is the inverse Laplace transform operator

$$P(s,t) = \mathcal{L}^{-1}u^{-1}\{I - \tilde{\psi}_d(u)\}G(s,u) \qquad [27]$$

in which $G(s,u)$ is the lattice Green's function matrix

$$G(s,u) = \frac{1}{N_1 \ldots N_d} \sum_{r_1=1}^{N_1} \cdots \sum_{r_d=1}^{N_d} \{\exp\,(-ik\cdot s)\}\{I - \tilde{\psi}(k,u)\}^{-1}. \qquad [28]$$

This function satisfies our required periodic boundary conditions (Eq. **2**). In the limit $N_\alpha \to \infty$ for all α,

$$G(s,u) = \frac{1}{(2\pi)^d}\int_{-\pi}^{\pi} \ldots \int_{-\pi}^{\pi} \{\exp\,(-ik\cdot s)\}\{I - \tilde{\psi}(k,u)\}^{-1} d^d k. \qquad [29]$$

We observe that, once given the matrix $\psi(s,\tau)$, Green's function $G(s,u)$ follows from Eq. **21** and the matrix $\psi_d(u)$ follows from Eq. **26**, so that all quantities that appear in our final formula, Eq. **29**, are given and the calculation of $P(s,t)$ is reduced to quadratures. Eq. **27** is the generalization of equations (V.11), (I.11), and (V.12) of ref. 5; there it was postulated that only one walker state exists in each cell.

We note that many interesting properties can be obtained directly from $\tilde{q}$. For example, in an infinite lattice the x-component of the mth moment is given by

$$\langle s_x{}^m(t)\rangle = \sum_s \sum_{j,j'} (s_x - s_x')^m P_{jj'}(s,s';t)p_{j'}$$
$$= \lim_{z\to 1,\ k\to 0} \mathcal{L}^{-1}\left\{(-i)^m \sum_{j,j'} (\partial^m \tilde{q}_{jj'}(k,u,z)/\partial k_x{}^m)u^{-1}[1 - \tilde{\psi}_j(u)]\right\}$$

in which p_j is the probability of initially being at state j. Equilibrium probabilities are given by

$$P_j = \lim_{t\to\infty} \sum_{s,m} P_{jm}(s,s',t)\, p_m$$
$$= \lim_{u,k\to 0,\ z\to 1} \sum_m \tilde{q}_{jm}(k,u,z)\,(1 - \tilde{\psi}_j(u))\,p_m.$$

GENERALIZED MASTER EQUATION FOR CONTINUOUS-TIME RANDOM WALK

We have constructed our required probability distributions by taking a weighted average over all paths available to the random walker. An alternative scheme for analyzing the subject walks is to construct a generalized master equation which is a differential equation to describe the evolution of the walk while it is in progress. We present an equation in the traditional form and show how the naturally appearing functions in the equation are related to those that characterize the individual paths.

With the identification established, one can discuss our continuous-time random walk in terms of a generalized master equation if one wishes.

We propose the following basic integral differential equation (generalized master equation) for $P_{jj'}(s,\tau)$:

$$\frac{dP_{jj'}(s,t)}{dt} = \int_0^t d\tau \sum_{j''s''} \{\Phi_{jj''}(s - s'',\tau)P_{j''j'}(s'',t - \tau) - \Phi_{j'',j}(s'' - s,\tau)P_{j,j'}(s,t - \tau)\}. \qquad [30]$$

Postulating a factorization analogous to Eq. **4a**, we set

$$\Phi_{j'',j}(s'' - s;\tau) \equiv F_{j'',j'}(s'' - s,\tau)\Phi_j(\tau) \qquad [31a]$$

in which the F function is precisely that given by Eq. **4a** and satisfies the normalization (Eq. **4b**). Then, using Eq. **4b**, we have

$$\Phi_j(\tau) = \sum_{j'',s''} \Phi_{j'',j}(s'' - s,\tau). \qquad [31b]$$

The independence of the sum on s is a consequence of the translational invariance of our lattice. We now show how the relaxation functions, $\Phi_j(\tau)$, are related to the waiting time functions, $\psi_j(\tau)$.

By taking Laplace and finite Fourier transforms of Eq. **30**

and Eq. **31** and using the initial conditions of Eq. **13**, we obtain

$$u\tilde{p}_{jj'}(k,u) - \delta_{jj'} = -\tilde{\phi}_j(u)\tilde{p}_{jj'}(k,u) + \sum_{j''} \tilde{\phi}_{jj''}(k,u)\tilde{p}_{j''j'}(k,u). \quad [32]$$

If we define $\tilde{\phi}_d(u)$ to be the diagonal matrix

$$\tilde{\phi}_d(u) = \begin{pmatrix} \tilde{\phi}_1(u) & 0 & . & . & 0 \\ 0 & \tilde{\phi}_2(u). & . & . & 0 \\ . & & . & . & . \\ 0 & 0 & . & . & \tilde{\phi}_m(u) \end{pmatrix}, \quad [33]$$

Eq. **32** has the matrix form

$$\{I - \tilde{\phi}(k,u)[uI + \tilde{\phi}_d(u)]^{-1}\}(uI + \phi_d(u))\tilde{p}(k,u) = I. \quad [34]$$

This equation can be made identical to Eq. **25** by equating the following two diagonal matrices:

$$[uI + \tilde{\phi}_d(u)] = u[I - \psi_d(u)]^{-1} \quad [35]$$

and

$$\tilde{\phi}(k,u)[uI + \tilde{\phi}_d(u)]^{-1} = \tilde{\psi}(k,u). \quad [36]$$

Consideration of the matrix elements involved in Eq. **35** implies that

$$\tilde{\phi}_j(u) = u\tilde{\psi}_j(u)/[1 - \tilde{\psi}_j(u)] \quad [37]$$

and

$$\tilde{\psi}_j(u) = \tilde{\phi}_j(u)/[u + \tilde{\phi}_j(u)]. \quad [38]$$

These expressions are the multistate generalizations of Equations 9a and 9b of ref. 13. (see also ref. 14). Eq. **36** is equivalent to

$$\tilde{\phi}(k,u) = u\tilde{\psi}(k,u)[I - \tilde{\psi}_d(u)]^{-1}. \quad [39]$$

The relationship between the components of $\tilde{\phi}$ and $\tilde{\psi}$ are then

$$\tilde{\phi}_{jl}(k,u) = u\tilde{\psi}_{jl}(k.u)/[1 - \tilde{\psi}_l(u)] \quad [40]$$

and

$$\tilde{\psi}_{jl}(k,u) = \tilde{\phi}_{jl}(k,u)/[u + \tilde{\phi}_l(u)]. \quad [41]$$

Eq. **37** and Eq. **38** are recovered by summing Eq. **40** and Eq. **41** over j, using Eq. **4a**, Eq. **4b**, and Eq. **31b.**

In conclusion, we have shown that a direct construction of the probability matrix, Eq. **27**, by a weighted sum over all achievable paths is equivalent to solving the generalized master equation, Eq. **30**, when the time-relaxed kernels of Eq. **30** are appropriately related to the transition rates through Eq. **40** and Eq. **41.**

An attractive feature of the generalized master equation is that it is precisely the form of the equation which has been derived (15–18) for the time development of the diagonal elements of the density matrix of a quantum mechanical system whose density matrix at time $t = 0$ is diagonal. In any special problem it is exceedingly difficult to derive the expression for $\Phi_{jj''}(\tau)$. However, by starting with Eq. **30** as a phenomenological equation to describe a process, one at least starts with an equation of the proper structure. Such an approach has been used recently (19) for the study of exciton transfer. The equivalence between the continuous-time random walk formalism and generalized master equations when there is no translational invariance, but also no internal states, has been noted.

The traditional approach to the generalized master equation has been to find wider sets of conditions that cause it to become equivalent to the Pauli master equation (or, if one is mathematically inclined, to the Kolmogorov equation for statistics of birth and death processes). Our attitude is to accept the fact that we have been given a very rich integral differential equation and marvel at the splendid variety of processes which it is capable of characterizing. For example, the generalized master equation describing transient photocurrents in certain xerographic films (4, 9) has a memory kernel decaying so slowly that the Pauli master equation is not valid in any limit.

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