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ON THE GENERAL RANDOM WALK FORMULATION FOR DIFFUSION IN MEDIA WITH MULTIPLE DIFFUSIVITIES

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Abstract

A general discrete multi-dimensional and multi-state random walk model is proposed to describe the phenomena of diffusion in media with multiple diffusivities. The model is a generalization of a two-state one-dimensional discrete random walk model (Hill [8]) which gives rise to the partial differential equations of double diffusion. The same partial differential equations are shown to emerge as a special case of the continuous version of the present general model. For two states a particular generalization of the model given in [8] is presented which is not restricted to nearest neighbour transitions. Under appropriate circumstances this two-state model still yields the partial differential equations of double diffusion in the continuum limit, but an example of circumstances leading to a radically different continuum limit is presented.

1. Introduction

After years of preoccupation with the classical diffusion process, physicists and engineers are now finding it increasingly necessary to adopt more sophisticated stochastic models to describe the subtle transport processes which arise in disordered materials [3, 11, 23, 24]. Sometimes the necessary tools have existed in the mathematical literature for some time. In some cases these have filtered through, while in others the wheel has been inadvertantly reinvented, a perhaps inevitable consequence of the current isolation [14] of "pure" mathematics from the physical sciences.

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In this paper we shall bring together, in an admittedly heuristic fashion, two currently popular developments of old themes in probability, *internal structure* and *divergent moments*. Our aim is to formulate a general system of equations describing, at a continuum level, random transport of charge, mass or energy under the action of several transport mechanisms of greatly differing efficiency. To do this, we shall examine a class of random walk processes on lattices (discrete spaces), in the limit as the lattice spacing and the time between steps approach zero simultaneously. In the manner of Landman, Montroll and Shlesinger [16, 22] and Hill, Liyanage and Gulati [8, 9, 18] and many others, we associate with each lattice site a number of internal states or degrees of freedom, so that there is random motion between states as well as motion from site to site. So long as simple moment conditions are obeyed, the equations of the continuum theory of multiple diffusion [1, 2, 8, 9, 18] are obtained. This is essentially a generalization of the familar central limit theorem. When these conditions are violated, for example with suitably long-ranged transition probabilities for individual displacements (*cf.* [6, 10, 13] in the ordinary random walk context), we obtain generalizations of the stable laws of Lévy [4, 7, 17].

In the following section we formulate the general discrete random walk model. In Section 3 we deduce the formal continuum limit of the model. The use of matrix formalism and transform methods enables us to deduce easily in Section 4 the differential equations of the simpler model of Hill [8], and to derive formulae for the mean displacement. In Section 5 we give a particular generalization of the model of [8] which is not restricted to nearest-neighbour transitions but may still give rise to the same double diffusion equations in the continuum limit. In Section 6 we give an example of this model involving the Riemann zeta function which does not give rise to the double diffusion equations and we examine in detail the various continuum limits. We have not attempted to formulate and prove rigorous theorems characterizing the continuum limit, but rather have limited ourselves to an at times heuristic investigation of the possible behaviours, especially in the case of divergent moments. (Some rigorous theorems of the central limit theorem type for systems with finite second moment have recently been proved by Krámli and Száz [15].)

2. General discrete formulation

We consider a class of spatially and temporally discretized multistate stochastic processes, namely random walks with internal states on periodic lattices. (These processes are known to probabilists under a variety of names, *e.g.* 'random walks controlled by a Markov chain'.) Specifically we consider a random walk taking place on the E dimensional simple cubic lattice ($E \geqslant 1$). If Δ is the lattice spacing, the allowed sites are the points $\mathbf{x} = \Delta \mathbf{l}$, where $\mathbf{l}$ is a vector with integer components. It is convenient initially to speak of the "lattice site $\mathbf{l}$", that is to suppress Δ and analyse the walk as if it took place on the set $\mathbf{Z}^E$. The length scale Δ is needed when we wish to take continuum limits in which both Δ and the time τ between successive steps are made to approach zero in a suitably constrained manner.

With each lattice site we associate m internal states. We have in mind the multiple diffusivity model [8, 9], in which each state corresponds to a distinct diffusivity. In the context of diffusion of ions in metals several high diffusivity paths can be identified such as dislocations, grain boundaries, free surfaces and internal microcracks. Similar equations arise in problems of cluster motion on surfaces [16, 22] and random walks on networks related to the Bethe lattice or Cayley tree [12].

Any step of the random walk may leave the walker in a new site, or a new state, or both, or he may pause at his present location (site and state). We define the transition probability law:

$$\Gamma_{nn'}(\mathbf{l} - \mathbf{l}') = \begin{cases} \text{probability of moving next to site } \mathbf{l} \text{ and} \\ \text{state } n, \text{ given that the walker is currently} \\ \text{at site } \mathbf{l}' \text{ in state } n'. \end{cases} \tag{2.1}$$

We note that implicit in this definition is that the random walk is translationally invariant. A more general model would have transition probability law $\Gamma_{nn'}(\mathbf{l}, \mathbf{l}') \neq \Gamma_{nn'}(\mathbf{l} - \mathbf{l}', 0)$ but this much harder problem is not considered here. We let $P_j(\mathbf{l})_n$ denote the probability that after j steps the walker is in state n at site $\mathbf{l}$ so that the evolution of the walk is described by

$$P_{j+1}(\mathbf{l})_n = \sum_{\mathbf{l}'} \sum_{n'=1}^{m} \Gamma_{nn'}(\mathbf{l} - \mathbf{l}') P_j(\mathbf{l}')_{n'}, \tag{2.2}$$

together with an initial condition which we take as

$$P_0(\mathbf{l})_n = U_n(\mathbf{l}) \qquad (n = 1, 2, \dots, m), \tag{2.3}$$

where $U_n(\mathbf{l}) \geqslant 0$ and

$$\sum_{\mathbf{l}} \sum_{n=1}^{m} U_n(\mathbf{l}) = 1. \tag{2.4}$$

We introduce matrix notation with $\Gamma(\mathbf{l} - \mathbf{l}')$ denoting an $m \times m$ matrix with nn'th element $\Gamma_{nn'}(\mathbf{l} - \mathbf{l}')$, and $\mathbf{P}_j(\mathbf{l})$ and $\mathbf{U}(\mathbf{l})$ being m dimensional column vectors with nth elements $P_j(\mathbf{l})_n$ and $U_n(\mathbf{l})$ respectively. Equations (2.2) and (2.3) become

$$\mathbf{P}_{j+1}(\mathbf{l}) = \sum_{\mathbf{l}'} \Gamma(\mathbf{l} - \mathbf{l}') \mathbf{P}_j(\mathbf{l}'), \tag{2.5}$$

$$\mathbf{P}_0(\mathbf{l}) = \mathbf{U}(\mathbf{l}). \tag{2.6}$$

The translational invariance of Γ allows us to exploit the discrete Fourier transform. If we define

$$\hat{\mathbf{P}}_j(\boldsymbol{\phi}) = \sum_{\mathbf{l}} e^{i\mathbf{l}\cdot\boldsymbol{\phi}} \mathbf{P}_j(\mathbf{l}), \tag{2.7}$$

then with $\hat{\Gamma}(\boldsymbol{\phi})$ and $\hat{\mathbf{U}}(\boldsymbol{\phi})$ defined analogously equations (2.5)and (2.6) become

$$\hat{\mathbf{P}}_{j+1}(\boldsymbol{\phi}) = \hat{\Gamma}(\boldsymbol{\phi})\hat{\mathbf{P}}_j(\boldsymbol{\phi}), \tag{2.8}$$

$$\hat{\mathbf{P}}_0(\boldsymbol{\phi}) = \hat{\mathbf{U}}(\boldsymbol{\phi}), \tag{2.9}$$

so that we have

$$\hat{\mathbf{P}}_j(\boldsymbol{\phi}) = \{\hat{\Gamma}(\boldsymbol{\phi})\}^j \hat{\mathbf{U}}(\boldsymbol{\phi}). \tag{2.10}$$

Now the inversion formula for the discrete Fourier transform is

$$\mathbf{P}_j(\mathbf{l}) = \frac{1}{(2\pi)^E} \int_B d^E\boldsymbol{\phi}\, e^{-i\mathbf{l}\cdot\boldsymbol{\phi}} \hat{\mathbf{P}}_j(\boldsymbol{\phi}), \tag{2.11}$$

where B denotes the first Brillouin zone, that is the hypercube of side 2π centred on $\boldsymbol{\phi} = \mathbf{0}$. Thus we deduce that

$$\mathbf{P}_j(\mathbf{l}) = \frac{1}{(2\pi)^E} \int_B d^E\boldsymbol{\phi}\, e^{-i\mathbf{l}\cdot\boldsymbol{\phi}} \{\hat{\Gamma}(\boldsymbol{\phi})\}^j \hat{\mathbf{U}}(\boldsymbol{\phi}), \tag{2.12}$$

and this equation is the formal solution of the random walk problem.

In applications we may only be interested in the site occupation probabilities rather than the distribution over the m states, so that we need to calculate

$$P_j(\mathbf{l}) = \sum_{n=1}^{m} P_j(\mathbf{l})_n = \boldsymbol{\omega}^T \mathbf{P}_j(\mathbf{l}), \tag{2.13}$$

with the row vector $\boldsymbol{\omega}^T = [1, 1, \ldots, 1]$ acting as a projection operator. From (2.12) the probabilities become

$$P_j(\mathbf{l}) = \frac{1}{(2\pi)^E} \int_B d^E\boldsymbol{\phi}\, e^{-i\mathbf{l}\cdot\boldsymbol{\phi}} \boldsymbol{\omega}^T \{\hat{\Gamma}(\boldsymbol{\phi})\}^j \hat{\mathbf{U}}(\boldsymbol{\phi}), \tag{2.14}$$

and clearly the willingness to accept only $P_j(\mathbf{l})$ rather than $\mathbf{P}_j(\mathbf{l})$ leads to no real simplications. We have now established the formal machinery to study the general problem of diffusion in media with multiple diffusivities. In the following section we consider the continuum limit.

3. The continuum limit

We define the probability density function for the walker, distributed over continuous space, at time $t = j\tau$ by

$$\mathbf{p}(\mathbf{x}, t) = \Delta^{-E} \mathbf{P}_j(\mathbf{l}), \tag{3.1}$$

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if **x** lies in the hypercube of side Δ centred on site **l** with its edges parallel to the coordinate axes. We also define

$$\hat{\mathbf{p}}(\mathbf{q}, t) = \hat{\mathbf{P}}_j(\Delta\mathbf{q}), \qquad \hat{\mathbf{u}}(\mathbf{q}) = \hat{\mathbf{U}}(\Delta\mathbf{q}), \tag{3.2}$$

for **q** within the hypercube of side $2\pi/\Delta$ centred on $\mathbf{q} = \mathbf{0}$. The notations $\hat{\mathbf{p}}$ and $\hat{\mathbf{u}}$ are used in anticipation of the fact that in the limit of Δ tending to zero $\hat{\mathbf{p}}$ is the continuous Fourier transform of **p**, since it follows from (2.11) in this limit that

$$\mathbf{p}(\mathbf{x}, t) = \frac{1}{(2\pi)^E}\int_{-\infty}^{\infty}\cdots\int d^E\mathbf{q}\, e^{-i\mathbf{x}\cdot\mathbf{q}}\hat{\mathbf{p}}(\mathbf{q}, t), \tag{3.3}$$

subject to modest hypotheses on the continuity of **p** and $\hat{\mathbf{p}}$.

On rewriting (2.8) in the form

$$\hat{\mathbf{P}}_{j+1}(\phi) - \hat{\mathbf{P}}_j(\phi) = -\{I - \hat{\Gamma}(\phi)\}\hat{\mathbf{P}}_j(\phi), \tag{3.4}$$

where I denotes the $m \times m$ identity matrix, we obtain with $t = j\tau$,

$$\hat{\mathbf{p}}(\mathbf{q}, t + \tau) - \hat{\mathbf{p}}(\mathbf{q}, t) = -\{I - \hat{\Gamma}(\Delta\mathbf{q})\}\hat{\mathbf{p}}(\mathbf{q}, t). \tag{3.5}$$

Thus provided that

$$A(\mathbf{q}) = \lim_{\Delta,\tau\to 0}\frac{\{I - \Gamma(\Delta\mathbf{q})\}}{\tau}, \tag{3.6}$$

exists for a suitable joint limit of Δ and τ we have formally established the equation

$$\frac{\partial\hat{\mathbf{p}}}{\partial t}(\mathbf{q}, t) = -A(\mathbf{q})\hat{\mathbf{p}}(\mathbf{q}, t). \tag{3.7}$$

This is a first order vector differential equation which we solve subject to the initial condition

$$\hat{\mathbf{p}}(\mathbf{q}, 0) = \hat{\mathbf{u}}(\mathbf{q}). \tag{3.8}$$

Consequently in terms of the matrix function $\exp(B)$ defined by the formal power series

$$\exp(B) = I + \sum_{k=1}^{\infty}\frac{1}{k!}B^k, \tag{3.9}$$

we have

$$\hat{\mathbf{p}}(\mathbf{q}, t) = \exp\{-tA(\mathbf{q})\}\hat{\mathbf{u}}(\mathbf{q}). \tag{3.10}$$

We observe that the spatial probability density for the walker with the distribution over internal states suppressed is

$$p(\mathbf{x}, t) = \boldsymbol{\omega}^T\mathbf{p}(\mathbf{x}, t), \tag{3.11}$$

which in Fourier transform space is given by

$$\hat{p}(\mathbf{q}, t) = \boldsymbol{\omega}^T\hat{\mathbf{p}}(\mathbf{q}, t). \tag{3.12}$$

Before proceeding to particular one-dimensional two-state examples in the following sections we make some general remarks. Firstly let the matrix $M(\mathbf{x}, t)$ denote the inverse Fourier transform of

$$\hat{M}(\mathbf{q}, t) = \exp\{-tA(\mathbf{q})\}. \tag{3.13}$$

Since the matrix $A(\mathbf{q})$ trivially commutes with itself we have for τ such that $0 \leqslant \tau \leqslant t$,

$$\hat{M}(\mathbf{q}, t) = \hat{M}(\mathbf{q}, t-\tau)\hat{M}(\mathbf{q}, \tau), \tag{3.14}$$

and therefore

$$M(\mathbf{x}, t) = \int_{-\infty}^{\infty} \cdots \int d^E\mathbf{y} M(\mathbf{x}-\mathbf{y}, t-\tau) M(\mathbf{y}, \tau). \tag{3.15}$$

We observe that equation (3.15) is the matrix form of the Bachelier-Chapman-Kolmogorov-Smoluchowski chain equation

$$P(\mathbf{x}, t) = \int_{-\infty}^{\infty} \cdots \int d^E\mathbf{y} P(\mathbf{x}-\mathbf{y}, t-\tau) P(\mathbf{y}, \tau), \tag{3.16}$$

for translationally invariant and temporally homogeneous Markov processes. (See for example Gikhman and Skorokhod [5], Chapter 7, and Mandl [19], or for a heuristic discussion, Montroll and West [20]. Solutions of equation (3.16) with modest smoothness properties are 'infinitely divisible distributions' [4, 7]. Lévy's stable distributions [17] defined below satisfy equation (3.16), and it has been erroneously stated [21] that they are its only solutions.)

Secondly we remark that for an arbitrary matrix-valued function $A(\mathbf{q})$, the vector $\mathbf{p}(\mathbf{x}, t)$ obtained from equation (3.10) will not necessarily be a valid probability density in the sense that its components are not necessarily positive or normalized for all τ, that is we may not have

$$\int_{-\infty}^{\infty} \cdots \int d^E\mathbf{x}\, \boldsymbol{\omega}^T \mathbf{p}(\mathbf{x}, t) = 1. \tag{3.17}$$

Conversely we need also observe that not every bonafide probability density $\mathbf{p}(\mathbf{x}, t)$ obtained from (3.10) is necessarily going to be found as the continuum limit of some multistate random walk. In the following section we show how the double diffusion equations arise from the above formulation.

4. Double diffusion equations

The double diffusion equations as formulated by Aifantis [1, 2] emerge from a two-state one-dimensional walk with transition probability law

$$\Gamma(l) = \begin{bmatrix} p_1 & 0 \\ 0 & p_2 \end{bmatrix} \delta_{l,1} + \begin{bmatrix} q_1 & 0 \\ 0 & q_2 \end{bmatrix} \delta_{l,-1} + \begin{bmatrix} r_1 & s_2 \\ s_1 & r_2 \end{bmatrix} \delta_{l,0}, \tag{4.1}$$

where $\delta_{i,j}$ denotes the usual Kronecker delta. If the particle is in state j ($j = 1,2$) it is assumed that it next moves one step to the right with probability p_j, one to the left with probability q_j, remains in position with probability r_j; each of these outcomes is assumed to take place without change of state. In addition it is assumed that the particle has probability s_j of switching to the other state without change of position. All probabilities are assumed independent of the position of the particle, and it is assumed that

$$p_j + q_j + r_j + s_j = 1 \qquad (j = 1,2), \tag{4.2}$$

i.e. there is no leakage of walkers from the system.

We find that

$$\hat{\Gamma}(\phi) = \begin{bmatrix} p_1 e^{i\phi} + q_1 e^{-i\phi} + r_1 & s_2 \\ s_1 & p_2 e^{i\phi} + q_2 e^{-i\phi} + r_2 \end{bmatrix}. \tag{4.3}$$

To obtain a finite continuum limit as the step length or lattice spacing Δ and the time τ between steps tend to zero, we assume that

$$p_j = \lambda^*(D_j + d_j\Delta), \quad q_j = \lambda^*(D_j - d_j\Delta), \quad s_j = \lambda^* k_j \Delta^2, \tag{4.4}$$

where d_j, D_j and k_j ($j = 1,2$) denote finite constants and λ^* denotes the finite positive limit of τ/Δ^2 as Δ and τ tend to zero. Writing $\phi = \Delta q$, we find that the function $A(q)$ defined by (3.6) becomes

$$A(q) = \begin{bmatrix} k_1 & -k_2 \\ -k_1 & k_2 \end{bmatrix} - iq \begin{bmatrix} 2d_1 & 0 \\ 0 & 2d_2 \end{bmatrix} + q^2 \begin{bmatrix} D_1 & 0 \\ 0 & D_2 \end{bmatrix}. \tag{4.5}$$

Thus from (3.7) utilizing the identities

$$\begin{aligned} \int_{-\infty}^{\infty} e^{iqx} \frac{\partial}{\partial x} \mathbf{p}(x,t)\, dx &= -iq\hat{\mathbf{p}}(q,t), \\ \int_{-\infty}^{\infty} e^{iqx} \frac{\partial^2}{\partial x^2} \mathbf{p}(x,t)\, dx &= -q^2\hat{\mathbf{p}}(q,t), \end{aligned} \tag{4.6}$$

(which hold unless $\mathbf{p}(x,t)$ is badly behaved as $|x| \to \infty$), wereadily obtain the second order matrix partial differential equation

$$\frac{\partial \mathbf{p}}{\partial t}(x,t) = \left\{ \begin{bmatrix} D_1 & 0 \\ 0 & D_2 \end{bmatrix} \frac{\partial^2}{\partial x^2} - \begin{bmatrix} 2d_1 & 0 \\ 0 & 2d_2 \end{bmatrix} \frac{\partial}{\partial x} - \begin{bmatrix} k_1 & -k_2 \\ -k_1 & k_2 \end{bmatrix} \right\} \mathbf{p}(x,t). \tag{4.7}$$

This is precisely the system of double diffusion equations given in [8].

In order to deduce means and variances from the above formulation we need to differentiate the exponential matrix function in (3.10). For example for the two-state one-dimensional vector moments we have

$$\mathbf{M}^{(k)} = \int_{-\infty}^{\infty} x^k \mathbf{p}(x,t)\, dx = (-i)^k \frac{\partial^k}{\partial q^k} \hat{\mathbf{p}}(q,t) \bigg|_{q=0}. \tag{4.8}$$

We illustrate these calculations for the mean only and moreover we determine only the scalar mean given by

$$M^{(1)} = \boldsymbol{\omega}^T \mathbf{M}^{(1)} = [1,1]^T \mathbf{M}^{(1)}. \tag{4.9}$$

Calculations for the higher vector moments are similar although the algebra becomes more complicated. It is convenient to introduce the constant matrices

$$D = \begin{bmatrix} D_1 & 0 \\ 0 & D_2 \end{bmatrix}, \quad V = \begin{bmatrix} 2d_1 & 0 \\ 0 & 2d_2 \end{bmatrix}, \quad J = \begin{bmatrix} k_1 & -k_2 \\ -k_1 & k_2 \end{bmatrix}, \tag{4.10}$$

so that we have

$$A(q) = J - iqV + q^2 D, \tag{4.11}$$

and we note the identity

$$\boldsymbol{\omega}^T J = \mathbf{0}, \tag{4.12}$$

which considerably simplifies the algebra.

From (3.10) and (4.8) we obtain

$$\mathbf{M}^{(1)} = -i\left\{\exp[-tA(0)]\frac{\partial \hat{\mathbf{u}}}{\partial q}(0) + \frac{\partial}{\partial q}\{\exp[-tA(q)]\}\bigg|_{q=0}\hat{\mathbf{u}}(0)\right\}. \tag{4.13}$$

Further from (3.9), (4.11) and (4.12) we find that

$$\boldsymbol{\omega}^T \exp[-tA(0)] = \boldsymbol{\omega}^T. \tag{4.14}$$

Moreover using

$$\frac{\partial}{\partial q}A(q)^k = \sum_{j=1}^{k} A(q)^{j-1}\left\{\frac{\partial}{\partial q}A(q)\right\}A(q)^{k-j}, \tag{4.15}$$

we can also deduce that

$$\boldsymbol{\omega}^T \frac{\partial}{\partial q}\exp[-tA(q)]\bigg|_{q=0} = -i\boldsymbol{\omega}^T V \sum_{k=1}^{\infty} \frac{(-t)^k}{k!} J^{k-1}. \tag{4.16}$$

But J is a singular matrix: for any vector $\left[\begin{smallmatrix} w_1 \\ w_2 \end{smallmatrix}\right]$ we have

$$J^n \begin{bmatrix} w_1 \\ w_2 \end{bmatrix} = (k_1 w_1 - k_2 w_2)(k_1 + k_2)^{n-1} \begin{bmatrix} 1 \\ -1 \end{bmatrix} \quad (n \geqslant 1). \tag{4.17}$$

From the above equations we may deduce that the scalar mean defined by (4.9) becomes

$$\begin{aligned} M^{(1)} = {} & -i\left\{\frac{\partial \hat{u}_1}{\partial q}(0) + \frac{\partial \hat{u}_2}{\partial q}(0)\right\} + 2t\{d_1 \hat{u}_1(0) + d_2 \hat{u}_2(0)\} \\ & + \frac{2(d_1 - d_2)[k_1 \hat{u}_1(0) - k_2 \hat{u}_2(0)]}{(k_1 + k_2)^2}\left\{1 - (k_1 + k_2)t - e^{-(k_1 + k_2)t}\right\}. \end{aligned} \tag{4.18}$$

For the case in which the random walker is initially localized at $l = 0$, *i.e.*

$$\mathbf{U}(l) = \begin{bmatrix} u_0 \\ v_0 \end{bmatrix} \delta_{l,0}, \quad \hat{\mathbf{u}}(q) = \begin{bmatrix} u_0 \\ v_0 \end{bmatrix}, \tag{4.19}$$

where $u_0 + v_0 = 1$, equation (4.18) reduces to

$$M^{(1)} = \frac{2(k_1 d_2 + k_2 d_1)t}{(k_1 + k_2)} + \frac{2(d_1 - d_2)(k_1 u_0 - k_2 v_0)}{(k_1 + k_2)^2}\left[1 - e^{-(k_1+k_2)t}\right]. \tag{4.20}$$

This result agrees with the continuum limit of an expression for the mean displacement of the two-state random walk given in [8].

5. Generalized double diffusion

For the two-state one-dimensional walk we let $p_j(n)$ $(j = 1,2)$ denote the probability of moving $|n|$ steps (to the right if n is a positive integer and to the left if n is a negative integer) and remaining in path j. With r_j and s_j as previously defined, so that path (state) changes can only occur when the walker stays at the same site, we have

$$\sum_{\substack{n=-\infty \\ n\neq 0}}^{\infty} p_j(n) + r_j + s_j = 1 \qquad (j = 1,2), \tag{5.1}$$

and the transition probability law is

$$\Gamma(l) = \sum_{\substack{n=-\infty \\ n\neq 0}}^{\infty} \begin{bmatrix} p_1(n) & 0 \\ 0 & p_2(n) \end{bmatrix} \delta_{l,n} + \begin{bmatrix} r_1 & s_2 \\ s_1 & r_2 \end{bmatrix} \delta_{l,0}. \tag{5.2}$$

We remark that the random walk model of the previous section results from taking $p_j(n)$ to be given by

$$p_j(n) = p_j \delta_{n,1} + q_j \delta_{n,-1} \qquad (j = 1,2). \tag{5.3}$$

We show below that the same double diffusion continuum limit also arises from (5.2) provided that

$$\mu_j = \sum_{n=-\infty}^{\infty} n p_j(n), \quad \eta_j = \sum_{n=-\infty}^{\infty} n^2 p_j(n) \qquad (j = 1,2), \tag{5.4}$$

are both finite and μ_j tends to zero in an appropriate manner with the step length Δ.

From (5.2), on making use of (5.1) we have

$$\hat{\Gamma}(\phi) = \begin{bmatrix} 1 - s_1 & s_2 \\ s_1 & 1 - s_2 \end{bmatrix} + \begin{bmatrix} \sum_{\substack{n=-\infty \\ n\neq 0}}^{\infty} p_1(n)(e^{in\phi} - 1) & 0 \\ 0 & \sum_{\substack{n=-\infty \\ n\neq 0}}^{\infty} p_2(n)(e^{in\phi} - 1) \end{bmatrix} \tag{5.5}$$

and since

$$\sum_{\substack{n=-\infty \\ n\neq 0}}^{\infty} p_j(n)(e^{in\phi} - 1) = i\phi\mu_j - \frac{\phi^2}{2}\eta_j + o(\phi^2), \tag{5.6}$$

we obtain

$$I - \hat{\Gamma}(\phi) = \begin{bmatrix} s_1 & -s_2 \\ -s_1 & s_2 \end{bmatrix} - i\phi\begin{bmatrix} \mu_1 & 0 \\ 0 & \mu_2 \end{bmatrix} + \frac{\phi^2}{2}\begin{bmatrix} \eta_1 & 0 \\ 0 & \eta_2 \end{bmatrix} + o(\phi^2). \tag{5.7}$$

Thus with

$$\mu_j = \lambda^* 2 d_j \Delta, \quad \eta_j = \lambda^* 2 D_j, \quad s_j = \lambda^* k_j \Delta^2, \tag{5.8}$$

where as previously λ^* denotes the finite positive limit of τ/Δ^2 as Δ and τ tend to zero, we obtain $A(q)$ given by (4.5) and therefore the same continuum limit (4.7) results.

If one or both of the individual path mean-square displacements per step η_j is infinite the preceding simple analysis is no longer applicable. Two distinct cases arise. In the first case, double diffusion equations can be derived if the continuum limit is modified. This case arises only when the series defining a divergent η_j is only very weakly divergent, in the sense that

$$\sum_{n=-\infty}^{\infty} p_j(n)[e^{in\phi} - 1] = i\mu_j\phi - \phi^2 L_j(\phi) + o(\phi^2), \tag{5.9}$$

with $L_j(\phi)$ 'slowly varying' in the sense of Feller [4], *i.e.* $L_j(\lambda\phi)/L_j(\phi) \to 1$ as $\phi \to 0$ for each $\lambda > 0$. So long as the functions $L_1(\phi)$and $L_2(\phi)$ are asymptotically proportional as $\phi \to 0$, double diffusion equations can be obtained in the continuum limit if the constraint $\tau \propto \Delta^2$ is replaced by $\tau \propto \Delta^2 L_j(\Delta)$. This case is a generalization to multistate processes of sharp forms of the central limit theorem which establish the existence of a Gaussian limit for sums of independent random variables with weakly divergent variances (see for example [4]).

In the second case, the series defining η_j diverges so strongly that the asymptotic form (5.9) is not obtained, and the usual double-diffusion limit is precluded. The following section is devoted to a detailed discussion of an example of such behaviour. We encounter what might be loosely described as multistate generalizations of the stable or Lévy distributions (see [4] or [7]), which arise from generalizations of the central limit theorem. A one-dimensional symmetric stable distribution of order μ has the probability density function $f_\mu(x)$ defined by

$$\hat{f}_\mu(q) = \int_{-\infty}^{\infty} e^{iqx} f_\mu(x)\, dx = \exp\left[-C|q|^\mu\right], \tag{5.10}$$

with C a positive constant. The parameter μ is constrained to the interval $0 < \mu \leqslant 2$, and the variance is finite only in the case $\mu = 2$, which is the Gaussian or normal distribution.

6. Example of generalized double diffusion

With $0 < \alpha_j < 2$ we consider the two-state random walk for which

$$p_j(n) = \frac{(1 - r_j - s_j)}{2\zeta(1 + \alpha_j)} \frac{1}{|n|^{1+\alpha_j}}, \qquad n \neq 0 \; (j = 1, 2); \tag{6.1}$$

here $\zeta(s)$ is the Riemann zeta function defined for $\mathrm{Re}(s) > 1$ by

$$\zeta(s) = \sum_{n=1}^{\infty} \frac{1}{n^s}. \tag{6.2}$$

Equation (6.1) defines a valid two-state random walk, but the lengths of individual steps have infinite variance.

We require the small ϕ behaviour of

$$\sum_{\substack{n=-\infty \\ n\neq 0}}^{\infty} p_j(n)[e^{in\phi} - 1] = \frac{1 - r_j - s_j}{\zeta(1 + \alpha_j)} \sum_{n=1}^{\infty} \frac{\cos n\phi - 1}{n^{1+\alpha_j}}. \tag{6.3}$$

For $0 < \alpha_j < 2$, it can be shown that as $\phi \to 0$,

$$\frac{1}{\zeta(1 + \alpha_j)} \sum_{n=1}^{\infty} \frac{\cos n\phi - 1}{n^{1+\alpha_j}} \sim \frac{-\pi|\phi|^{\alpha_j}}{2\zeta(1 + \alpha_j)\Gamma(1 + \alpha_j)\sin(\pi\alpha_j/2)}. \tag{6.4}$$

(See, e.g. Gillis and Weiss [6].) When $0 < \alpha_j < 2$, a formal contour integral argument based on the Mellin transform [10] may be used to establish the complete asymptotic expansion

$$\frac{1}{\zeta(1 + \alpha_j)} \sum_{n=1}^{\infty} \frac{\cos n\phi - 1}{n^{1+\alpha_j}} \sim \frac{-\pi|\phi|^{\alpha_j}}{2\zeta(1 + \alpha_j)\Gamma(1 + \alpha_j)\sin(\pi\alpha_j/2)} + \sum_{n=1}^{\infty} \frac{(-1)^n \zeta(1 + \alpha_j - 2n)\phi^{2n}}{(2n)!\zeta(1 + \alpha_j)}. \tag{6.5}$$

(The argument can be made rigorous when $1/2 < \alpha_j < 2$.)

The presence of both a series in integer powers of ϕ and a nonanalytic component (here proportional to $|\phi|^{\alpha_j}$) in the series (6.3), revealed in (6.5), is to be expected from experience with single-state random walks having long-ranged transitions [10, 11, 13]. When $0 < \alpha_j < 2$, the nonanalytic component dominates the power series as $\phi \to 0$, and the qualitative behaviour of the multistate random walk is sensitive to the asymptotic behaviour of the probability distributions $p_j(n)$.

For $\alpha_j > 2$ there is still a nonanalytic component in the series (6.3), but it no longer dominates as $\phi \to 0$, and so the results of Section 5 are applicable. In the borderline case $\alpha_j = 2$, the dominant term is proportional to $|\phi|^2 \log|\phi|$, giving an example of a 'weakly divergent' moment as discussed at the end of Section 5. A double diffusive limit is therefore obtained in the case $\alpha_1 = \alpha_2 = 2$, so long as Δ and τ are constrained such that $\tau \propto \Delta^2 \log(1/\Delta)$.

We now examine more closely the consequences of assuming that $0 < \alpha_j < 2$ $(j = 1, 2)$. From (5.5), (6.3) and (6.4) we may show that

$$I - \hat{\Gamma}(\phi) = \begin{bmatrix} s_1 & -s_2 \\ -s_1 & s_2 \end{bmatrix} + \begin{bmatrix} (1 - r_1 - s_1) K_1 |\phi|^{\alpha_1} & 0 \\ 0 & (1 - r_2 - s_2) K_2 |\phi|^{\alpha_2} \end{bmatrix}$$
$$+ o\left(|\phi|^{\alpha_1}\right) + o\left(|\phi|^{\alpha_2}\right), \tag{6.6}$$

where the constants K_j $(j = 1, 2)$ are defined by

$$K_j = \frac{\pi}{2\zeta(1 + \alpha_j)\Gamma(1 + \alpha_j)\sin(\pi\alpha_j/2)}. \tag{6.7}$$

For this example a wide variety of continuum limits are possible. Without loss of generality we suppose that $\alpha_1 \leqslant \alpha_2$. A well defined continuum limit is only possible when $s_1 = O(\Delta^{\alpha_1})$ and $s_2 = O(\Delta^{\alpha_1})$. We examine the following two situations for Δ tending to zero:

(i) $s_1 = o(\Delta^{\alpha_1})$, $s_2 = o(\Delta^{\alpha_1})$,

(ii) $s_1 \sim k_1 \Delta^{\alpha_1}$, $s_2 \sim k_2 \Delta^{\alpha_1}$;

in the second of these cases it is assumed that at least one of the nonnegative constants k_1 and k_2 is nonzero.

For *case* (i) with $\tau \sim \Delta^{\alpha_1}$ as Δ tends to zero we find from (6.6)

$$A(q) = \begin{cases} (1 - r_1) K_1 |q|^{\alpha_1} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} & \text{if } \alpha_1 < \alpha_2, \\ |q|^{\alpha_1} \begin{bmatrix} (1 - r_1) K_1 & 0 \\ 0 & (1 - r_2) K_1 \end{bmatrix} & \text{if } \alpha_1 = \alpha_2, \end{cases} \tag{6.8}$$

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so that from (3.10) we obtain

$$\hat{\mathbf{p}}(q,t) = \begin{cases} \begin{bmatrix} \exp\left[-t(1-r_1)K_1|q|^{\alpha_1}\right]\hat{u}_1(q) \\ \hat{u}_2(q) \end{bmatrix} & \text{if } \alpha_1 < \alpha_2, \\ \begin{bmatrix} \exp\left[-t(1-r_1)K_1|q|^{\alpha_1}\right]\hat{u}_1(q) \\ \exp\left[-t(1-r_2)K_1|q|^{\alpha_1}\right]\hat{u}_2(q) \end{bmatrix} & \text{if } \alpha_1 = \alpha_2. \end{cases} \tag{6.9}$$

Thus for case (i) two possibilities arise for the continuum limit. We either have a stable law confined to one path (when $\alpha_1 < \alpha_2$) or a stable law of order α_1 for each state with no changes of state (if $\alpha_1 = \alpha_2$).

For *case* (ii) with $\tau \sim \Delta^{\alpha_1}$ we find

$$A(q) = \begin{cases} \begin{bmatrix} k_1 & -k_2 \\ -k_1 & k_2 \end{bmatrix} + (1-r_1)K_1|q|^{\alpha_1}\begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} & \text{if } \alpha_1 < \alpha_2, \quad (6.10\text{a}) \\ \begin{bmatrix} k_1 & -k_2 \\ -k_1 & k_2 \end{bmatrix} + |q|^{\alpha_1}\begin{bmatrix} (1-r_1)K_1 & 0 \\ 0 & (1-r_2)K_2 \end{bmatrix} & \text{if } \alpha_1 = \alpha_2. \quad (6.10\text{b}) \end{cases}$$

It is clear that in general for case (ii) the ability to change state is retained in the continuum limit. In very special cases some simplification occurs. If the two matrices in (6.10b) commute, $\exp[-tA(q)]$ can be factorized. The necessary and sufficient conditions for commutation are that either (a) $k_1 = k_2 = 0$ (which is just case (i)) or (b) $(1-r_1)K_1 = (1-r_2)K_2 = \lambda$ say and $\alpha_1 = \alpha_2$. In the latter case we find that the distribution over space and between states is given by

$$\hat{\mathbf{p}}(q,t) = \exp\left(-t\lambda|q|^{\alpha_1}\right)\exp\left\{-t\begin{bmatrix} k_1 & -k_2 \\ -k_1 & k_2 \end{bmatrix}\right\}\hat{\mathbf{u}}(q). \tag{6.11}$$

More generally, if we are only interested in which state the walker occupies, we may obtain a vector $\boldsymbol{\sigma}(t)$, with components $\sigma_j(t)$ the probability of being in state j at time t, by setting $q = 0$ in (6.10). We find that

$$\begin{bmatrix} \sigma_1(t) \\ \sigma_2(t) \end{bmatrix} = \exp\left\{-t\begin{bmatrix} k_1 & -k_2 \\ -k_1 & k_2 \end{bmatrix}\right\}\hat{\mathbf{u}}(0). \tag{6.12}$$

It is easily verified that $\sigma_1(t) + \sigma_2(t) = 1$, corresponding to conservation of probability. Using (4.17), it is easy to show that

$$\begin{bmatrix} \sigma_1(t) \\ \sigma_2(t) \end{bmatrix} = \begin{bmatrix} \hat{u}_1(0) \\ \hat{u}_2(0) \end{bmatrix} + \frac{[k_1\hat{u}_1(0) - k_2\hat{u}_2(0)]}{(k_1+k_2)}\left[e^{-(k_1+k_2)t} - 1\right]\begin{bmatrix} 1 \\ -1 \end{bmatrix}, \tag{6.13}$$

and as $t \to \infty$ we find an 'equilibrium distribution' between the two states,

$$\begin{bmatrix} \sigma_1(t) \\ \sigma_2(t) \end{bmatrix} \sim \begin{bmatrix} k_2/[k_1 + k_2] \\ k_1/[k_1 + k_2] \end{bmatrix}, \tag{6.14}$$

which is insensitive to the values of α_1 and α_2.

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