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Dispersion of Particles in Periodic Media

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We discuss the long-time properties of the dispersion of particles in periodic media, using the random walk formalism. Exact asymptotic results are obtained for the average velocity and the diffusion coefficient, expressed in terms of the Green's function of the random walk inside the periodically repeated unit cell. We explicitly calculate the transport coefficients for several specific cases of interest, including a system with "dead zones," a simple model for field-induced trapping, and a one-dimensional map leading to deterministic diffusion.

KEY WORDS: Random walks; periodic systems; transport coefficients.

1. INTRODUCTION

The transport or dispersion of neutrally buoyant particles in fluids is governed by two mechanisms: molecular diffusion and flow convection. Diffusion plays an important role in a wide variety of physical and chemical processes, such as chemical reactions, mixing of fluids, spreading of pollutants, chromatography, and electrophoresis. It is therefore of fundamental and practical importance to understand the interaction between these two mechanisms, i.e., how does the flow pattern affect the dispersion of passive particles, and what is the resulting concentration profile of the particles?

Although the enhancement of dispersion by turbulence is well known, the fact that even laminar flow can increase the dispersion is far less well known. In 1953 Taylor[(1)] showed that the longitudinal dispersion of particles suspended in a Poiseuille flow in a cylindrical tube of radius R is

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described by an effective diffusion coefficient D(D is a measure for the width of the concentration profile of the particles) which is given by

$$D = D_m + \frac{\langle v \rangle^2 R^2}{48 D_m} \tag{1}$$

where D_m is the molecular diffusion coefficient and $\langle v \rangle$ is the average flow velocity. The molecular diffusion coefficient is typically of the order of 10^{-5}–10^{-8} cm^2/sec in liquids, and so the contribution of the flow to the effective dispersion of particles is by far the dominant effect.

The study of convection-induced dispersion in a general velocity field is very complicated. However, analytic results can be obtained for an important subclass of flows, namely the ones with a periodic velocity field. Examples are flow profiles arising as a consequence of hydrodynamic instabilities, such as the Rayleigh–Bénard system and the circular Couette system, or flow profiles in periodic media.[2,3] In these cases, even when the mean velocity $\langle v \rangle$ of the flow is zero, the dispersion of the particles occurs through the combination of convection along the streamlines and molecular diffusion between the streamlines. In particular, for the Rayleigh–Bénard instability, Sagues and Horsthemke[4] found (using a perturbative method) an effective transport coefficient D equal to

$$D = D_m + \frac{\langle v^2 \rangle d^2}{(a^2 + \pi^2) D_m} \tag{2}$$

The convection-induced contribution is again dominant, and has a form similar to that encountered in the original Taylor problem [see Eq. (1)].

Different approaches to describe such systems have been proposed in the literature (see, e.g., refs. 2–9). For the applications that we have in mind, we will restrict ourselves to the simple case in which the motion of the particles can be described as a Markovian random walk across a system consisting of a periodically repeated unit cell (see Fig. 1). In this case, it can be shown that the long-time concentration profile converges to a Gaussian form,[3] and it suffices to calculate the long-time convection velocity and the long-time diffusion coefficient of the particles. We have evaluated these quantities using the method of the moments, introduced by Aris[11] in the context of Taylor dispersion phenomena, but identical results can be obtained in the context of the multistate random walk formalism.[7] The unit cell contains a number of states or lattice sites between which the particles can jump according to prescribed jump rates. In the applications to be discussed here, these sites represent the spatial locations at which the

[3] A proof can be given along lines similar to those of ref. 10.

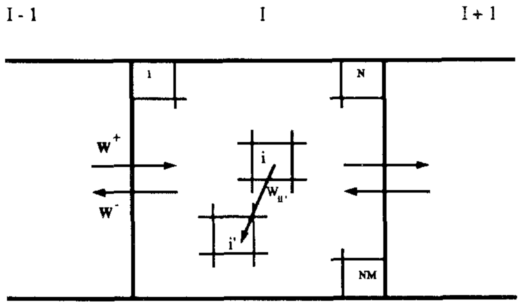

Fig. 1. Periodically repeated unit cell consisting of $N \times M$ states.

particles reside, and jumping from one state to another thus constitutes a physical displacement of the particle. This is the way in which processes mentioned earlier can be modeled, e.g., Taylor dispersion or the dispersion of particles in a Rayleigh–Bénard flow. It is, however, also possible that the sites represent different physical states of the particle, such as the geometrical conformation of a molecule or its charge, and these affect the transition rates of the random walk that it performs. This description can be used to model processes such as chromatography, electrophoresis, NMR, molecular rotational dynamics, etc.

Our main purpose in this paper will be to calculate explicitly the asymptotic convection and diffusion coefficients for several applications of the above-described model. These include dispersion in spatially periodic flows (cf. the discussion given above), the effect of "dead zones,"[12,13] dispersion enhancement in field-induced trapping,[14–17] a one-dimensional chaotic map leading to deterministic diffusion,[18,19] and a new general result for Taylor dispersion.[20,21]

2. EXACT ASYMPTOTIC RESULTS

The periodically repeated unit cell consists of a rectangular array of $N \times M$ lattice sites or states such as represented in Fig. 1. For simplicity, we will assume that the lattice sites inside the unit cell are at a distance 1 of each other. The length of the unit cell along the x axis is thus equal to N, and the position of a particle is given by $x(t) = NI(t)$, where $I(t)$ is the index of the unit cell that is occupied at time t. We have disregarded here the spatial variation on the particle's position inside the unit cell since this correction is negligible in the long-time limit. As the particles move through the system, they jump from one state to another. The jump rate to go from a state i to another state i' (not necessarily a nearest neighbor) inside the same unit cell is given by the matrix element $W_{i'i}$, while the transitions from state i to state i' in the previous or next unit cell are denoted by the matrix elements $W^-_{i'i}$ and $W^+_{i'i}$, respectively. The probability to find a particle in state i in unit cell I at time t is the solution of the following master equation:

$$\begin{aligned}\partial_t P(i, I, t) = \sum_{i'} \{ &[W_{ii'} P(i', I, t) - W_{i'i} P(i, I, t)] \\ &+ [W^-_{ii'} P(i', I+1, t) - W^-_{i'i} P(i, I, t)] \\ &+ [W^+_{ii'} P(i', I-1, t) - W^+_{i'i} P(i, I, t)]\}\end{aligned} \tag{3}$$

with the sum over i' running over all the sites in the unit cell. One can show that in the long-time limit, the probability profile for the position of the particle converges to a Gaussian law[10] and consequently only the first two moments $\langle x(t) \rangle = N\langle I(t) \rangle$ and $\langle x^2(t) \rangle = N^2 \langle I^2(t) \rangle$ are needed to characterize this behavior. The equations governing the time evolution of these moments can easily be derived from Eq. (3) using the method of moments[11] and can be solved by Laplace transformation. To formulate the results, it is convenient to introduce the following stochastic matrix $\mathbf{T}$ $(\sum_i T_{ij} = 0)$:

$$\begin{cases} T_{ij} = W_{ij} + W^+_{ij} + W^-_{ij}, & i \neq j \\ T_{ii} = W_{ii} + W^+_{ii} + W^-_{ii} - \sum_j (W_{ji} + W^+_{ji} + W^-_{ji}) & \end{cases} \tag{4}$$

and its Green's function

$$\mathbf{G}(s) = \frac{1}{s\mathbf{1} - \mathbf{T}} \tag{5}$$

Since we will be concerned with the long-time properties of the transport, we will only need the small-s expansion of the Green's function:

$$G_{ij}(s) \underset{s \to 0}{=} \frac{P_i^{\mathrm{st}}}{s} - G^1_{ij} + O(s) \tag{6}$$

Note that the stochastic matrix $\mathbf{T}$ describes a random walk inside the unit cell with periodic boundary conditions, which does not depend on the value of the "diagonal" jump frequencies W^+_{ii} and W^-_{ii}. After a straightforward calculation, we find the following asymptotic results for the drift velocity v and the diffusion coefficient D:

$$v = \lim_{t \to \infty} \frac{\langle x(t) \rangle}{t} = N \sum_l \sum_m (W^+_{lm} - W^-_{lm}) P^{\mathrm{st}}_m \tag{7}$$

$$D = \lim_{t \to \infty} \frac{\langle x^2(t)\rangle - \langle x(t)\rangle^2}{2t}$$
$$= N^2 \left[\frac{1}{2} \sum_l \sum_m (W_{lm}^+ + W_{lm}^-) P_m^{st} \right.$$
$$\left. - \sum_l \sum_m \sum_n \sum_r (W_{lm}^+ - W_{lm}^-) G_{mn}^1 (W_{nr}^+ - W_{nr}^-) P_r^{st} \right] \tag{8}$$

where the summations run over all the $N \times M$ sites of the unit cell. We conclude that the average velocity v and the effective diffusion coefficient D can be expressed in terms of $\mathbf{P}^{st}$ and $\mathbf{G}^1$; thus, only the small-s behavior of the Green's function of the random walk on the unit cell with periodic boundary conditions is needed. In the next section we will apply these results to specific cases of interest.

3. APPLICATIONS

3.1. Rotating Fluid Rolls

As a first example, we consider the transport of particles moving through a system of rotating rolls. The unit cell modeling this situation has four states (see Fig. 2a) with transition rates $k(1-g)$ between nearest neighbors in the counterclockwise direction and $k(1+g)$ in the clockwise direction. The particles can move through the boundaries to the next or previous cells with jump rates k. The matrices $\mathbf{T}$ (with periodic boundary conditions), $\mathbf{W}^+$, and $\mathbf{W}^-$ are given by

$$\mathbf{T} = \begin{pmatrix} -3k & k(2-g) & k(1+g) & 0 \\ k(2+g) & -3k & 0 & k(1-g) \\ k(1-g) & 0 & -3k & k(2+g) \\ 0 & k(1+g) & k(2-g) & -3k \end{pmatrix} \tag{9}$$

$$\mathbf{W}^+ = \begin{pmatrix} 0 & k & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad \mathbf{W}^- = \begin{pmatrix} 0 & 0 & 0 & 0 \\ k & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & k & 0 \end{pmatrix} \tag{10}$$

The average velocity v equals zero and D [Eq. (8)] reduces to

$$D = k\left(1 + \frac{g^2}{g^2+2}\right) \tag{11}$$

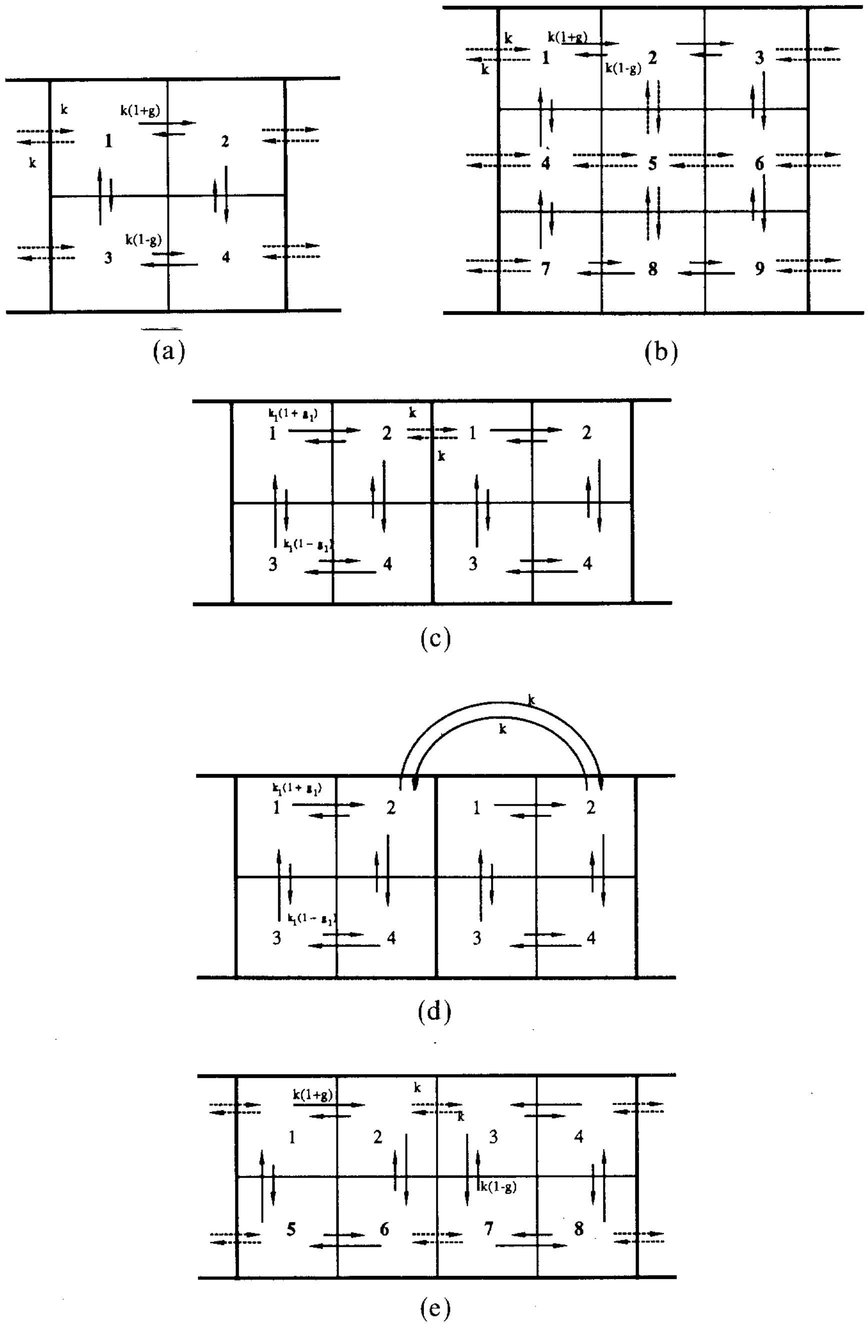

Fig. 2. Unit cells for systems with rotating fluid rolls.

In an analogous way the result for a unit cell with nine states (see Fig. 2b) is obtained as

$$D = k\left(1 + \frac{2g^2(g^2+11)}{g^4+13g^2+18}\right) \tag{12}$$

Two variants of the above-described system illustrate the surprising modifications introduced by other types of transitions between neighboring unit cells. In the first case (Fig. 2c), the particles leave the unit cell only by jumping from state 1 to state 2 and vice versa. In this case, the average velocity equals zero and one obtains for the diffusion coefficient

$$D = k\left(1 - \frac{3+g^2}{5+7g^2}\right) \tag{13}$$

Whereas in the previous examples [cf. Eq. (11)] the presence of the rotating fluid rolls tends to enhance the dispersion, we now find that the dispersion is reduced.

On the other hand, if particles can only jump from state 2 to the same state in the neighboring unit cells (Fig. 2d), we recover a result identical to that for the "plain" one-dimensional system, i.e., $v=0$ and $D=k$.

The Rayleigh–Bénard flow pattern is characterized by counterrotating fluid rolls. To model this situation, we considered a very simple system in which the unit cell contains eight states (see Fig. 2e). The average velocity v of the dispersed particles is again zero, while D is given by

$$D = k\left(1 + \frac{g^2}{g^2+4}\right) \tag{14}$$

Note that the particles are more effectively dispersed in the case where the fluid rolls have the same direction [cf. Eq. (11)] than in the case of alternating rolls.

3.2. Dead Pockets

The presence of "dead zones" can significantly modify the long-term dispersion of the particles.[13,22] To investigate this effect in more detail, we will study some representative examples. We start with the unit cell shown in Fig. 3a. It has two states between which the particles can switch with jump rates $k_1(1 \pm g_1)$. One of these states corresponds to the "dead pocket," while in the other state the particle can move "freely" along the

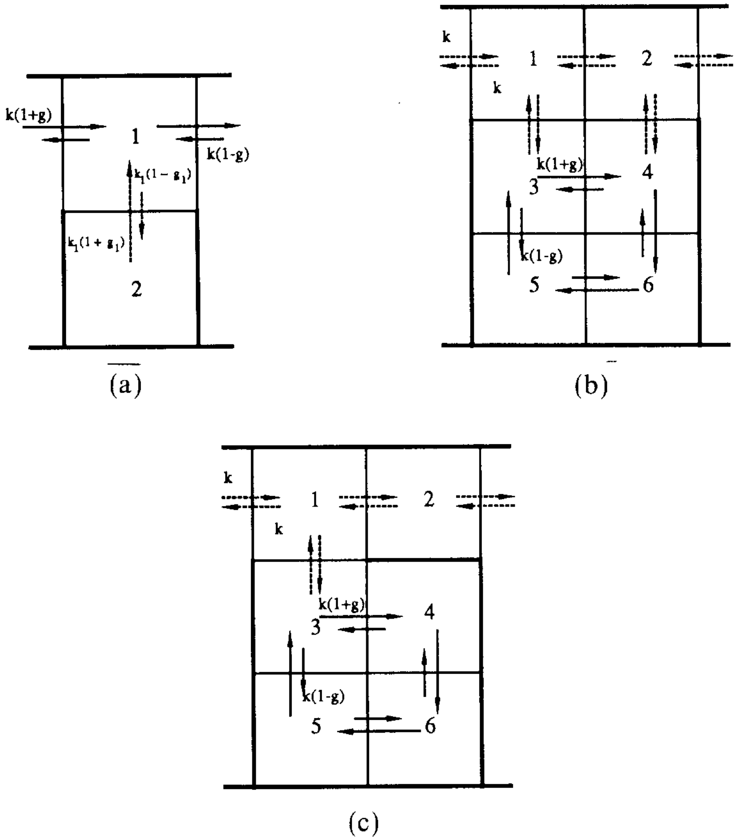

Fig. 3. Systems with "dead pockets."

horizontal direction. The velocity v and the diffusion coefficient D for this example are given by

$$v = kg(1 + g_1) \tag{15}$$

and

$$D = \frac{k}{2}(1 + g_1)\left(1 + \frac{k}{k_1}g^2(1 - g_1)\right) \tag{16}$$

which are to be contrasted with the results $2kg$ and k, respectively, for a biased random walk in the absence of a "dead pocket" (corresponding to the case $g_1 = 1$). In particular, for $g_1 = 0$, we find that the effect of the "dead pocket" is to reduce the convection velocity by a factor 2, while the diffusion coefficient is modified by a factor $\frac{1}{2}(1+g^2(k/k_1))$. Consequently, dispersion is dramatically enhanced in the limit $k_1 \to 0$.

In the system represented in Fig. 3b, we have introduced a "dead pocket" modeling a trapped vortex. We find that $v=0$ and

$$D=\frac{k}{3}\left(1+\frac{2k_1(1+g_1^2)}{k(3+g_1^2)+10k_1(1+g_1^2)}\right) \tag{17}$$

D is thus restricted to the following interval:

$$\frac{k}{3} \leqslant D \leqslant 2\frac{k}{5} \tag{18}$$

Remarkably, the addition of a reflecting boundary between the states two and four (Fig. 3c) completely modifies this result and leads to a diffusion coefficient independent of k_1 and g_1:

$$D=\frac{k}{3} \tag{19}$$

3.3. Dispersion Enhancement in Field-Induced Trapping

Consider a random walk on a line, with transition probabilities to the right and to the left equal to $k(1+g)$ and $k(1-g)$, respectively. In this simple situation, the drift velocity increases monotonically with the strength of the bias $v=2kg$, while the asymptotic dispersion coefficient does not depend on the applied bias, $D=k$. The situation changes drastically in the presence of dead-end branches. An external bias now has a dual effect: on the one hand, the bias induces convection in the direction of the field, while on the other hand, dead ends that point in the same direction as the field are converted into traps. In this case, one observes that the convection speed v goes through a maximum as a function of the applied bias. This phenomenon has been discussed in the context of conduction on the percolating cluster and models have been proposed to describe it.[14–17] The point that we want to emphasize here is that the dispersion coefficient also behaves in a nontrivial way: it also goes through a maximum as a function of the applied bias. To illustrate this behavior, we have plotted in Figs. 4a and 4b the values of v and D as a function of the bias g, for a periodic, one-dimensional system with a dead-end branch of depth 3. The corresponding unit cell is represented in Fig. 4c. Note that the maximum for both v and D as a function of g becomes more pronounced and shifts to larger values

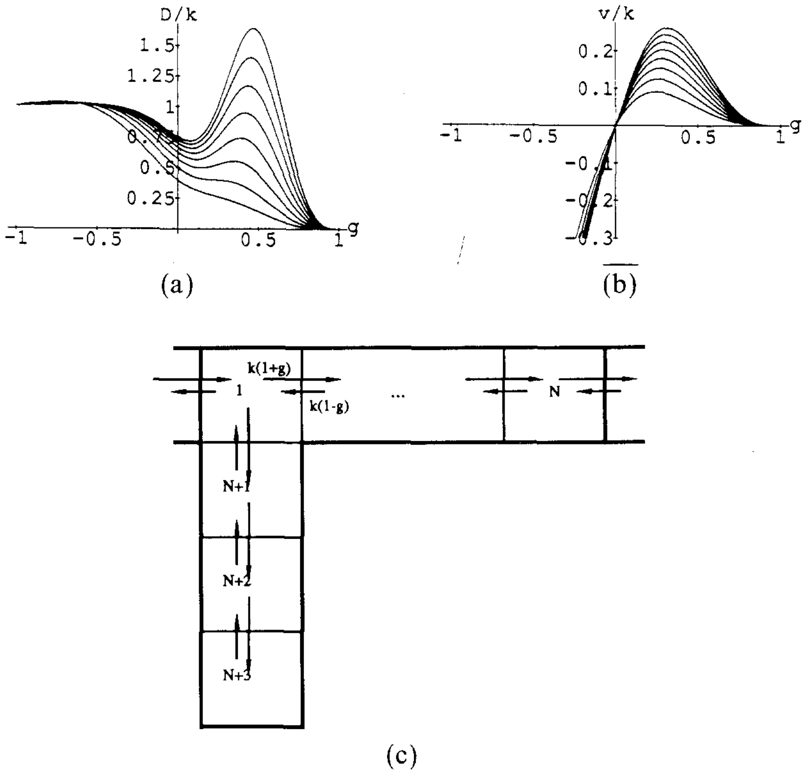

Fig. 4. (a) D/k and (b) v/k as a function of g and for increasing length N of the unit cell ($N=1$, bottom curves, up to $N=8$), and (c) model used to illustrate the effect of field-induced trapping.

of the bias as the densities of traps decreases (or as the length N of the unit cell increases). We expect a similar behavior for a random distribution of traps.

3.4. Diffusion Induced by a Deterministic Map

Diffusion is an ubiquitous example of an irreversible, dissipative process. Numerous attempts have been undertaken to derive it in a rigorous way from deterministic, reversible microscopic dynamics. A very attractive deterministic model that gives rise to "deterministic diffusion" was introduced some time ago by Geisel and Nierwetberg.[18] Here we present a few simple variants of this model, which, moreover, have the advantage that they can be mapped exactly onto a random walk with a periodically repeated unit cell.[19] Consider first the piecewise linear one-dimensional map represented in Fig. 5a. Note that particles can "escape" from a unit cell I, located at $[I, I+1]$, to the cells $I+1$ and $I-1$ through the sections A_2 and A_4, respectively. Furthermore, the intervals A_1 through A_5 provide a natural Markovian partition, since the endpoints of these intervals map into each other. Finally, it is clear that if the probability density is piecewise constant on these intervals at time 0, it will remain so for all times. Consequently, for such initial conditions, the dynamic evolution induced by the map is equivalent to a Markovian random walk process of the type that we have been discussing in this paper, with the important difference that time is a discrete variable here. As discussed in detail in ref. 23, having a time-discrete rather than a time-continuous random walk leaves the drift velocity unchanged but reduces the diffusion coefficient by an amount proportional to the time step Δt and the square of the drift velocity:

$$D_{\text{discrete time}} = D_{\text{continuous time}} - \frac{v^2}{2} \Delta t \tag{20}$$

For the above-described map, the time step Δt is equal to 1 and the unit cell is schematically represented in Fig. 5b. The transition matrix reads

$$\mathbf{T} = \begin{pmatrix} -1+a_1 & 1 & a_1 & 0 & a_1 \\ a_2 & -1 & a_2 & 0 & a_2 \\ a_3 & 0 & -1+a_3 & 0 & a_3 \\ a_4 & 0 & a_4 & -1 & a_4 \\ a_5 & 0 & a_5 & 1 & -1+a_5 \end{pmatrix} \tag{21}$$

with

$$a_1 + a_2 + a_3 + a_4 + a_5 = 1 \tag{22}$$

The following results are obtained for v and D [cf. Eqs. (7), (8), and (20)]:

$$v = \frac{a_2 - a_4}{1 + a_2 + a_4} \tag{23}$$

$$D = \frac{a_2 + 3a_2^2 + 4a_2^2 a_4 + 2a_2 a_4 + 4a_2 a_4^2 + 3a_4^2 + a_4}{2(1 + a_2 + a_4)^3} \tag{24}$$

For the most symmetrical case with the values of the parameters a_1, a_2, a_3, a_4, and a_5 chosen as

$$a_1 = a_3 = a_5 = \tfrac{1}{4} \qquad \text{and} \qquad a_2 = a_4 = \tfrac{1}{8} \tag{25}$$

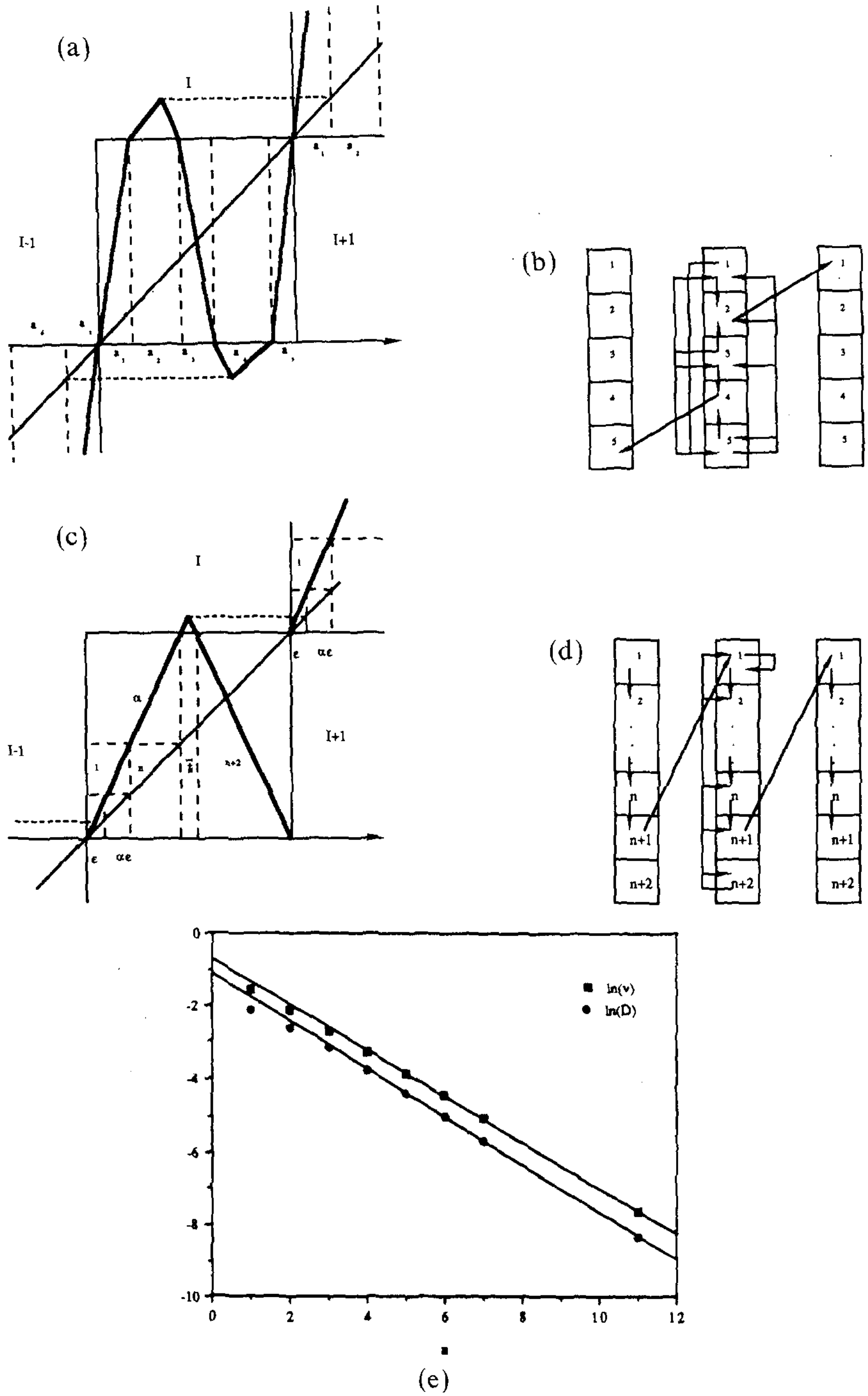

Fig. 5. (a, c) Examples of deterministic maps that give rise to diffusion, (b, d) the corresponding unit cells, (e) and the results for v and D.

one finds that $D = 1/10$. On the other hand, the Lyapunov exponent of this map is equal to $\lambda = \ln 4$, while the Sinai–Kolmogorov entropy characterizing the trajectories that never escape the unit is equal to $H_{SK} = \ln 3$. As is well known,[24] the probability to leave the unit cell per iteration, which we denote by $2k$ since a particle can move to both the previous and the next cell, can be expressed in terms of these quantities as $\ln(1-2k) = H_{SK} - \lambda$, hence $k = 1/8$. The connection with the resulting transport coefficient D is not so clear in the present case.[25] Indeed, the details of the reinjection mechanism of a particle into the neighboring cell are needed to determine the value of D. In particular, it is found in the present case that $D = 1/10$ is not equal to k (which is the result for an unbiased random walk with hopping rate k).

An interesting variant of the above model, displaying a combination of convection and diffusion, is represented in Figs. 5c and 5d. Note that this model is characterized by the parameter n ($n = 1, 2,...$) such that the escape from the unit cell becomes increasingly difficult as n becomes larger. At the same time, the number of states of the Markovian partition increases as $n+2$. The slope α of the map is chosen such that the particles leaving the unit cell from state $n+1$ are injected uniformly into partition 1 of the next unit cell

$$\alpha = 2(1+2\varepsilon) \tag{26}$$

Additionally, in order for the nth image of partition 1 to map onto the unit interval, ε must satisfy

$$\alpha^n \varepsilon = 1 \tag{27}$$

This fully determines ε for every value of n. The results for D and v for increasing values of n are shown in Fig. 5e, while the asymptotic for n large read

$$v \underset{n\to\infty}{\sim} 2^{-n} \tag{28}$$

$$D \underset{n\to\infty}{\sim} 2^{-n} \tag{29}$$

3.5. Taylor Dispersion Phenomena

As a last application, we turn to the description of a layered system in which the particles perform nearest-neighbor random walks inside each layer as well as between the layers (see Fig. 6). The unit cell contains $1 \times M$ states, labeled by $i = 1,..., M$, with transitions between state i and its nearest

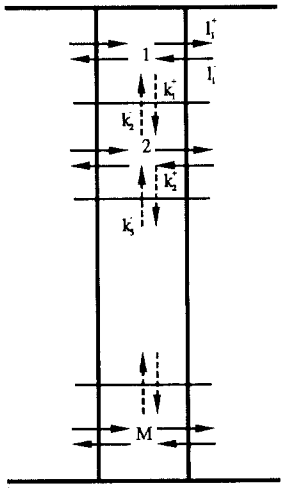

Fig. 6. Unit cell used to describe Taylor dispersion phenomena.

neighbors $i-1$ and $i+1$ occurring at rates k_i^- and k_i^+, respectively. We assume that the particles cannot leave the system through the upper or the lower boundaries; hence the jump rates k_1^- and k_M^+ are equal to zero. The transition matrix **W** is given by

$$W=\begin{pmatrix} -k_1^+ & k_2^- & 0 & \cdots & 0 & 0 \\ k_1^+ & -(k_2^+ + k_2^-) & k_3^- & 0 & \cdots & 0 \\ 0 & k_2^+ & & \ddots & & \vdots \\ \vdots & & & & k_{M-1}^- & 0 \\ 0 & \cdots & 0 & k_{M-2}^+ & -(k_{M-1}^+ + k_{M-1}^-) & k_M^- \\ 0 & 0 & \cdots & 0 & k_{M-1}^+ & -k_M^- \end{pmatrix} \tag{30}$$

while the matrices $\mathbf{W}^+$ and $\mathbf{W}^-$ read

$$W_{ij}^+ = \delta_{ij} l_i^+ \tag{31}$$

$$W_{ij}^- = \delta_{ij} l_i^- \tag{32}$$

Using a recursive calculation outlined in ref. 21, we find the following analytic result for the long-time behavior of the Green's function:

$$P_i^{\mathrm{st}} = \frac{1}{\mathcal{N}} k_1^+ \cdots k_{i-1}^+ k_{i+1}^- \cdots k_M^- \tag{33}$$

with

$$\mathcal{N} = k_2^- \cdots k_M^- + k_1^+ k_3^- \cdots k_M^- + \cdots + k_1^+ \cdots k_{M-1}^+ \tag{34}$$

and

$$G_{ij}^1 = -\sum_{r=1}^{M-1} \sum_{l=1}^{r} \sum_{n=1}^{r} P_i^{\text{st}} \frac{(\delta_{in} - P_n^{\text{st}})(\delta_{jl} - P_l^{\text{st}})}{k_r^+ P_r^{\text{st}}} \tag{35}$$

The average velocity v is obtained from Eq. (7),

$$v = \sum_{i=1}^{M} (l_i^+ - l_i^-) P_i^{\text{st}} \tag{36}$$

and the effective diffusion coefficient D from Eq. (8),

$$D = \frac{1}{2} \sum_{i=1}^{M} (l_i^+ + l_i^-) P_i^{\text{st}} + \sum_{r=1}^{M-1} \frac{1}{k_r^+ P_r^{\text{st}}} \left[\sum_{i=1}^{M} \sum_{n=1}^{r} (l_i^+ - l_i^-) P_i^{\text{st}} (\delta_{in} - P_n^{\text{st}}) \right]^2 \tag{37}$$

In the particular case of two layers with $l_1^+ = l_2^- = l_1$ and $l_1^- = l_2^+ = l_2$, Eqs. (36) and (37) reduce to the following simple results ($k_1^+ = k_1$ and $k_2^- = k_2$):

$$v = \frac{(k_1 - k_2)(l_2 - l_1)}{k_1 + k_2} \tag{38}$$

$$D = \frac{l_1 + l_2}{2} + 4k_1 k_2 \frac{(l_1 - l_2)^2}{(k_1 + k_2)^3} \tag{39}$$

The results given in Eqs. (36) and (37) are the generalizations to the case of a discrete system of those given in refs. 20 and 21. The latter results are recovered by taking the following continuum limit (introducing the parameter a, the width of the unit cell):

$$\begin{cases} a \to 0 \\ I \to \infty \\ l_i^{\pm} \to \infty \end{cases} \quad \text{with} \quad \begin{cases} x = Ia \\ a^2 \dfrac{(l_i^+ + l_i^-)}{2} = D_i \\ a(l_i^+ - l_i^-) = v_i \end{cases} \quad \text{fixed} \tag{40}$$

One thus finds

$$v = \sum_{i=1}^{M} v_i P_i^{\text{st}} \tag{41}$$

$$D = \sum_{i=1}^{M} D_i P_i^{\text{st}} + \sum_{r=1}^{M-1} \frac{(\sum_{l=1}^{r} (v_l - v) P_l^{\text{st}})^2}{k_r^+ P_r^{\text{st}}} \tag{42}$$

4. CONCLUSION

We have shown that the calculation of the effective diffusion coefficient for the dispersion of particles in spatially periodic systems can be reduced to the calculation of the Green's function for the random walk in the periodically repeated unit cell. In the case of small unit cells, the Green's function can be calculated explicitly (possibly with the use of symbolic manipulators like Macsyma). In other cases, such as a random walk in a one-dimensional unit cell with general transition rates, the small-s expansion of the Green's function is known analytically. In all these models, the interplay between convection and molecular diffusion leads to a modification of the transport properties that can be calculated analytically. Using this formalism, we have discussed several models that display or illustrate phenomena likely to occur in more realistic systems.

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