Physica 132A (1985) 253–268
North-Holland, Amsterdam

ON THE CALCULATION OF RANDOM WALK PROPERTIES FROM LATTICE BOND ENUMERATION

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Received 6 March 1985

We examine the range of validity of previously developed lattice bond enumeration methods for the calculation of asymptotic properties of random walks on inhomogeneous periodic lattices.

1. Introduction

Some time ago, Shuler[1]) developed an Ansatz, hereafter referred to as the lattice bond enumeration method (LBE), for the calculation of asymptotic properties of nearest-neighbor random walks on so-called sparsely periodic lattices. In the case of a two-dimensional square lattice, such a sparsely periodic lattice is obtained by periodically removing a number of vertical columns. An example is given in fig. 1. On this lattice, motion in the y-direction is only possible at a subset of points, the so-called intersection sites (we assume that only steps to nearest neighbors are taken). The jump probabilities are $\frac{1}{4}$ in each possible direction at an intersection site, and $\frac{1}{2}$ at a non-intersection site. More generally, suppose that the lattice has periodicity k, i.e. there is one vertical column for every kth site along the horizontal direction. If $\langle n_x(n)\rangle$ and $\langle n_y(n)\rangle$ are the average number of steps in the x- and y-direction after a total number of n steps, then Shuler's Ansatz is

$$\frac{\langle n_x(n)\rangle}{\langle n_y(n)\rangle} \simeq \frac{B_x}{B_y} = k \quad (n \to \infty). \tag{1.1}$$

Here B_x and B_y are the number of bonds in the x- and y-direction within a unit cell of the lattice, or rather within an irreducible lattice fragment (ILF), which is

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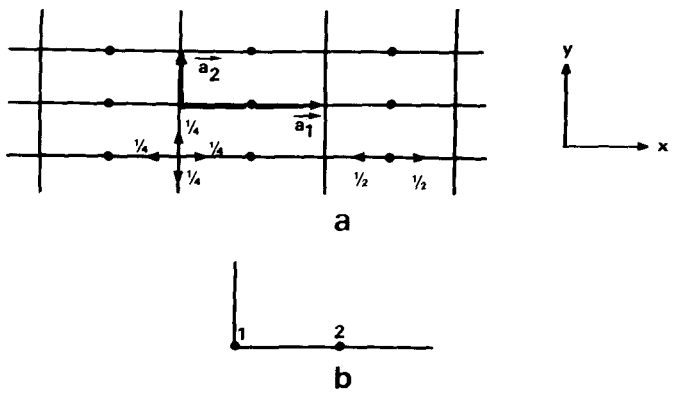

Fig. 1. (a) Sparsely periodic lattice with horizontal periodicity $k_x = 2$ and vertical periodicity $k_y = 1$. The arrows indicate the allowed steps of the walker. The corresponding jump probabilities are indicated. (b) Irreducible lattice fragment belonging to fig. 1a.

the smallest lattice fragment from which the lattice can be constructed by repetition (see fig. 1b). Since the walk is *locally unbiased**, the variances of the displacement are given by (we assume the lattice constants are unity)

$$\langle x^2(n)\rangle = \langle n_x(n)\rangle \simeq n\,\frac{k}{k+1} \quad (n \to \infty) \tag{1.2a}$$

and

$$\langle y^2(n)\rangle = \langle n_y(n)\rangle \simeq n\,\frac{1}{k+1} \quad (n \to \infty)\,, \tag{1.2b}$$

where we used that $\langle n_x(n)\rangle + \langle n_y(n)\rangle = n$. These results have been confirmed, among others, by the matrix method for calculating the moments of multistate random walks developed recently[2]). A comparison between a periodic and a random distribution of vertical columns was subsequently made in ref. 3, where it was shown that if a column is present with probability $p = k^{-1}$, and absent with probability $q = 1 - p$, then the result for the mean square displacements is identical to (1.2) with probability 1. In other words, the spatial arrangement of the columns is irrelevant; only the average density k^{-1} is important. For a discussion of another Ansatz made by Shuler[1]) to calculate the probability of return to the origin and the expected number of distinct sites visited, see refs. 2 and 3.

* A locally unbiased walk is one in which the single step averages of the displacement from any site on the lattice in every space direction are zero.

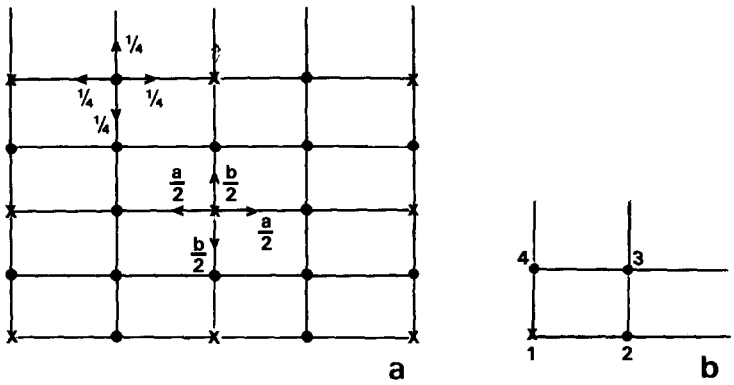

Fig. 2. (a) Two-dimensional anisotropic lattice. x scattering sites. The stepping probabilities are as indicated. (b) Irreducible lattice fragment belonging to fig. 2a.

A slightly modified lattice bond enumeration method (MLBE) was developed by Shuler and Mohanty[4]) for random walks on lattices with anisotropic scatterers. An example is given in fig. 2. They obtained the ratio of the mean square displacements in the x- and y-direction as

$$\frac{\langle x^2(n)\rangle}{\langle y^2(n)\rangle} \simeq \frac{\hat{B}_x}{\hat{B}_y} \quad (n \to \infty), \tag{1.3}$$

where $\hat{B}_x(\hat{B}_y)$ is obtained by counting all bonds in the ILF which are parallel to the $x(y)$-axis, and multiplying each bond by the corresponding stepping probability. For the example of fig. 2, this method yields (the ILF consists of 4 sites, labeled 1–4 in fig. 2b)

$$\frac{\langle x^2(n)\rangle}{\langle y^2(n)\rangle} \simeq \frac{a/2+3/4}{b/2+3/4} \quad (n \to \infty), \tag{1.4}$$

a result which is confirmed in section 3 below. In ref. 3 it was found that for these types of lattices the spatial arrangement of the scatterers is relevant, i.e. the ratio $\langle x^2(n)\rangle/\langle y^2(n)\rangle$ can be different for different arrangements with the same number of scatterers in the corresponding ILF's.

It is the purpose of this paper to clarify why the lattice bond enumeration methods give the correct results in the cases discussed above, and to find necessary and/or sufficient conditions for their validity. We will show that the range of validity of the LBE and MLBE is quite limited. This implies no restriction whatsoever on the applicability of the general formalism developed in ref.

2, since it is precisely this method by which the results of the present paper are derived.

For later use we briefly summarize the matrix method of ref. 2 for the mean square displacements of locally unbiased walks. The position of the random walker on an inhomogeneous periodic lattice, i.e. a lattice consisting of a periodically repeated unit cell with internal sites, (the lattices of figs. 1 and 2 are special cases) is labeled by a vector $\boldsymbol{l}$, indicating the unit cell which the walker occupies, and an index α, which denotes the site (or "internal state") within the unit cell which the walker occupies. All Greek indices run from 1 to m, where m is the number of sites in the unit cell. Fundamental in our method is the stochastic matrix $\mathbf{T}$ (i.e. $\Sigma_\alpha T_{\alpha\beta} = 1$) with elements

$$T_{\alpha\beta} = \sum_{\boldsymbol{l}} T_{\alpha\beta}(\boldsymbol{l}), \tag{1.5}$$

where $T_{\alpha\beta}(\boldsymbol{l} - \boldsymbol{l}')$ is the transition probability of unit cell $\boldsymbol{l}'$ and internal state β to unit cell $\boldsymbol{l}$ and internal state α. We always assume that the walk is *irreducible*, i.e. each site $(\boldsymbol{l}, \alpha)$ can be reached from every other site $(\boldsymbol{l}', \beta)$ after a sufficient number of steps. The basic quantity to be determined is the right eigenvector $\boldsymbol{\pi}$ of $\mathbf{T}$, corresponding to the maximal eigenvalue $\lambda_0 = 1$. Let $\boldsymbol{r}(n)$ be the displacement of the walker after n steps, with components $r_i(n) \equiv \boldsymbol{r}(n) \cdot \boldsymbol{e}_i$, where $\{\boldsymbol{e}_i\}$ is a complete set of unit vectors spanning the Euclidean space $\mathbb{R}^d$. Then the following result for the covariances of the displacement was derived in ref. 2, *assuming that the walk is locally unbiased*:

$$\langle r_i(n) r_j(n) \rangle \simeq 2nD_{ij} \quad (n \to \infty), \tag{1.6}$$

where the brackets $\langle \cdots \rangle$ denote the average over all realizations of the random walk and where the diffusion coefficients D_{ij} are given by

$$D_{ij} = \tfrac{1}{2} \sum_{\beta} \langle r_i r_j \rangle_\beta \pi_\beta . \tag{1.7}$$

Here the single-step covariance from state β is defined as

$$\langle r_i r_j \rangle_\beta \equiv \sum_{\boldsymbol{r}} r_i r_j p_\beta(\boldsymbol{r}), \tag{1.8}$$

with $p_\beta(\boldsymbol{r})$ the single-step transition probability from the internal state β.

It turns out that for some walks which are not locally unbiased the Ansatz (1.1) is still valid, although no corresponding statement for the variances holds. In order to discuss this we need an asymptotic result for the *average number of steps* in the various directions $\{\boldsymbol{e}_i\}$, not merely the corresponding variances of the displacement. This is done in appendix A. In order to state the result, let us label the collection of transition probabilities $\{T_{\alpha\beta}(\boldsymbol{l})\}$ in the ILF for different α and $\boldsymbol{l}$ and a *given* value of β by a parameter ν_β. To each value of $\nu_\beta = 1, 2, \ldots$ corresponds a certain transition from a site $(\boldsymbol{0}, \beta)$ to a site $[\boldsymbol{l}(\nu_\beta), \alpha(\nu_\beta)]$. For example, in the lattice of fig. 1 there are four different transitions from an intersection site 1 and two from a non-intersection site 2, so ν_1 runs from 1 to 4 and ν_2 runs from 1 to 2. The probability of the transition labeled by ν_β is indicated by p_{ν_β}, and the corresponding displacement by $\boldsymbol{r}^{(\nu_\beta)}$.* Then the average number $\langle n_{\nu_\beta}(n)\rangle$ of steps of type ν_β after n steps has the large-n behavior (see appendix A)

$$\langle n_{\nu_\beta}(n)\rangle \simeq p_{\nu_\beta}\pi_\beta n \quad (n\to\infty), \tag{1.9}$$

where π_β is the asymptotic occupation probability of the site β from which the transition of type ν_β occurs. If we know $\langle n_{\nu_\beta}(n)\rangle$ for all $\{\nu_\beta\}$ then the average number of steps in the direction $\boldsymbol{e}_i$ can be obtained by summing over all those values of $\{\nu_\beta\}$ which correspond to transitions in the direction $\boldsymbol{e}_i$.

In the following we first discuss the LBE in section 2 and then in section 3 the MLBE.

2. Validity of the LBE

The result (1.2) can be obtained from the more general formula (3.4) of ref. 3 by putting $p_1 = q_1 = \frac{1}{4}$; $q_2 = q_3 = \cdots = q_k = 0$. From that formula it is also evident that the Ansatz (1.1) by which the result is found by bond counting no longer applies for general values of $\{p_i, q_i\}$. In the following we derive some sufficient conditions for the validity of the LBE.

We assume the following conditions, besides irreducibility of the multistate random walk:

i) all transitions are parallel to one of the axes $\{\boldsymbol{e}_i\}$ of a d-dimensional lattice; a transition with zero displacement (i.e. a pause) is also allowed;

ii) for any transition between different sites also the reverse transition occurs (not necessarily with the same probability);

iii) only transitions to nearest neighbors are allowed;

iv) all lattice constants are unity.

* So $p_{\nu_\beta} \equiv p_\beta[\boldsymbol{r}^{(\nu_\beta)}]$, where $p_\beta(\boldsymbol{r})$ is the same as in eq. (1.8).

Assumptions (i)–(iii) are necessary in order that the LBE has unambiguous meaning, whereas (iv) is assumed for convenience.

The ratio of the average numbers of steps in two directions $\boldsymbol{e}_i$ and $\boldsymbol{e}_j$ after n steps is given by

$$\frac{\langle n_i(n)\rangle}{\langle n_j(n)\rangle} = \frac{\sum_{\{\nu_\beta\}}^{(i)} \langle n_{\nu_\beta}(n)\rangle}{\sum_{\{\nu_\beta\}}^{(j)} \langle n_{\nu_\beta}(n)\rangle} \simeq \frac{\sum_{\{\nu_\beta\}}^{(i)} p_{\nu_\beta}\pi_\beta}{\sum_{\{\nu_\beta\}}^{(j)} p_{\nu_\beta}\pi_\beta} \quad (n\to\infty), \tag{2.1}$$

where (1.9) has been used. The summations are over all values of $\nu_1, \nu_2, \ldots, \nu_m$, i.e. over all transitions from the sites in the ILF. However, the symbol $\Sigma^{(i)}$ means that only transitions in the direction $\boldsymbol{e}_i$ should be taken into account [and similarly for $\Sigma^{(j)}$].

If the following condition is met:

$$p_{\nu_\beta}\pi_\beta = c \quad (\beta = 1, 2, \ldots, m), \tag{2.2}$$

where c is a constant, then clearly

$$\frac{\langle n_i(n)\rangle}{\langle n_j(n)\rangle} \simeq \frac{\sum_{\{\nu_\beta\}}^{(i)}}{\sum_{\{\nu_\beta\}}^{(j)}} = \frac{B_i}{B_j}. \tag{2.3}$$

Here B_i and B_j are the number of bonds in the ILF parallel to $\boldsymbol{e}_i$ and $\boldsymbol{e}_j$, respectively. Hence under the condition (2.2) the Ansatz (1.1) holds. In the case of the sparsely periodic lattice, the relation (2.2) is indeed satisfied [see eq. (2.2.33) of ref. 2 for the equilibrium probabilities $\{\pi_\beta\}$ for this case].

Before discussing a somewhat more general situation in which (2.2) holds, let us briefly discuss the implications of (2.2) for the variances of the displacement in the directions $\boldsymbol{e}_i$ and $\boldsymbol{e}_j$. If the walk is *locally unbiased*, then

$$\frac{\langle r_i^2(n)\rangle}{\langle r_j^2(n)\rangle} = \frac{\sum_{\{\nu_\beta\}} [r_i^{(\nu_\beta)}]^2 \langle n_{\nu_\beta}(n)\rangle}{\sum_{\{\nu_\beta\}} [r_j^{(\nu_\beta)}]^2 \langle n_{\nu_\beta}(n)\rangle}, \tag{2.4}$$

where $\boldsymbol{r}^{(\nu_\beta)}$ is the displacement during a transition of type ν_β. Under the assumptions (i)–(iv) above, $r_i^{(\nu_\beta)} = 1$ if a transition is parallel to the direction $\boldsymbol{e}_i$

and zero otherwise. Hence, if (2.3) holds,

$$\frac{\langle r_i^2(n)\rangle}{\langle r_j^2(n)\rangle} = \frac{\langle n_i(n)\rangle}{\langle n_j(n)\rangle} \simeq \frac{B_i}{B_j} \quad (n \to \infty)\,. \tag{2.5}$$

So if (2.2) is satisfied, the ratio of the variances can be obtained by the LBE as well, provided the walk is locally unbiased.

Remarks

i) The LBE will remain valid if (2.2) is only satisfied for those transitions ν_β for which $r^{(\nu_\beta)}$ is not identically zero [if $r^{(\nu_\beta)}$ is zero, this corresponds to a "pause" of the walker]. Although the numerators and denominators in (2.5) will be affected, the corresponding ratios are not.

ii) Although (2.2) looks like a detailed balance relation between the transitions among the m internal states, it is in fact stronger, for detailed balance affirms (2.2) only for those transitions which lead from an internal state β to a *different* internal state α. For example, in the lattice of fig. 2 of ref. 3, detailed balance was satisfied, but (2.2) is not true in that case and neither is (2.3).

iii) One should distinguish between the LBE as applied to the ratio of *average numbers of steps*, as in (2.3), and the LBE as applied to the ratio of variances, as in (2.5). For example in the case of the modified sparsely periodic lattice discussed in subsection 3.3 of ref. 3, the LBE is correct as far as the numbers of steps is concerned [this follows from the lemma below] but gives the wrong result for the ratio of the variances. The reason is that this walk is not locally unbiased in the vertical direction. In fact, the LBE predicts the same result for all arrangements of bonds with the same density of bonds in the various space directions. In the case of the modified sparsely periodic lattice the vertical variance depends upon the arrangement, however[3]).

The next question which comes up is under what conditions eq. (2.2) is satisfied. One of the cases in which this occurs is when the walker at each site i selects *with equal probability* n_i^{-1} one of the open pathways leading away from the site i presently occupied, where n_i is the total number of open pathways from site i [this is the "*myopic ant*" algorithm of ref. 5]. The proof is based on the following lemma, which is proved in appendix B:

Lemma. Consider an irreducible (not necessarily nearest neighbor) random walk on an inhomogeneous periodic lattice. Assume that for every transition $i \to j$ between two sites i and j also the reverse transition $j \to i$ is allowed. Let the probability of pausing at a site $i = (l', \beta)$ be t_β and let the probability of any transition $i \to j$ with $i \neq j$ be equal to $(1 - t_\beta)$, multiplied by the inverse of the total number n_β of transitions originating from site $i = (l', \beta)$ and ending in a different site $j = (l, \alpha)$.

Then the eigenvector $\boldsymbol{\pi}$ of the matrix $\mathbf{T}$ corresponding to this multistate random walk has components

$$\pi_\alpha = cn_\alpha(1 - t_\alpha)^{-1} \quad (\alpha = 1, 2, \ldots, m), \tag{2.6}$$

where

$$c = \left[\sum_{\beta=1}^{m} n_\beta(1 - t_\beta)^{-1}\right]^{-1}.$$

Corollary. Since $p_{\nu_\beta} = n_\beta^{-1}(1 - t_\beta)$ for all transitions which do not correspond to a pause of the walker, eq. (2.2) is satisfied for all ν_β with $\boldsymbol{r}^{(\nu_\beta)} \neq \boldsymbol{0}$. This implies the validity of eq. (2.3) [see remark (i) above].

Another situation in which (2.2) is satisfied at least for $\boldsymbol{r}^{(\nu_\beta)} \neq \boldsymbol{0}$ is when both p_{ν_β} is constant for all $\{\nu_\beta\}$ with $\boldsymbol{r}^{(\nu_\beta)} \neq \boldsymbol{0}$ and π_β is constant for all β. [The latter requires that $\pi_\beta = m^{-1}$, i.e. that the matrix $\mathbf{T}$ is symmetric*.] This is the case, for example, for the "*blind ant*" algorithm of ref. 5, where the walker chooses with a *constant* probability n^{-1} one of the n_i open pathways leading away from a site i and pauses at site i with probability $(1 - n_i n^{-1})$, where $n_i \leq n$. This leads to the same ratio (2.3) as the "myopic ant" algorithm.

As an application, consider a sparsely periodic 2-D lattice with a unit cell of $m \times m$ sites, where a number of *both* columns *and* rows is deleted, so that m_1 columns and m_2 rows remain. Then four types of sites occur, type 1 with two horizontal and two vertical bonds, type 2 with two horizontal bonds and no vertical bonds, and type 3 with two vertical and no horizontal bonds. Let us use the "myopic ant" algorithm, i.e. the jump probability along each of the bonds is $\frac{1}{4}$ for a site of type 1 and $\frac{1}{2}$ for the other types. Then the *lemma* is applicable and the walk is locally unbiased, so from (2.5)

$$\frac{D_x}{D_y} = \frac{B_x}{B_y} = \frac{m \cdot m_2}{m \cdot m_1} = \frac{m_2}{m_1}$$

and since for a locally unbiased nearest neighbor walk without pauses and steps of unit length $D_x + D_y = \frac{1}{2}$, we also have

$$D_x = \frac{1}{2}\frac{m_2}{m_1 + m_2}, \quad D_y = \frac{1}{2}\frac{m_1}{m_1 + m_2}.$$

* A symmetric stochastic matrix is also called "doubly stochastic" since both column and row sums are unity.

The formulae (1.2a) and (1.2b) are recovered by putting $m_2 = k$, $m_1 = 1$. Again, the arrangement of the rows and columns is irrelevant. This implies that the case of random arrangements is also very simple. For, if the probability of a column or row to be present is p and q, respectively, independent of the other rows or columns, then by the same method as in section 3 of ref. 3 we find that, with probability one,

$$D_x = \frac{1}{2}\frac{q}{p+q}, \quad D_y = \frac{1}{2}\frac{p}{p+q}.$$

3. Validity of the MLBE

To investigate the range of validiy of formula (1.3) for lattices with anisotropic scatterers, we first write down the general expression for the ratio D_x/D_y of the diffusion coefficients, which is given by (1.7) as

$$\frac{D_x}{D_y} = \frac{\sum_\beta \langle x^2 \rangle_\beta \pi_\beta}{\sum_\beta \langle y^2 \rangle_\beta \pi_\beta}. \tag{3.1}$$

This formula would certainly give the same result as (1.3) if the occupation probabilities $\{\pi_\beta\}$ of all the sites were equal. This would be equivalent to saying that the matrix $\mathbf{T}$ is doubly stochastic. However, the matrix $\mathbf{T}$ for the case of fig. 2 is given by

$$\mathbf{T} = \begin{bmatrix} 0 & \frac{1}{2} & 0 & \frac{1}{2} \\ a & 0 & \frac{1}{2} & 0 \\ 0 & \frac{1}{2} & 0 & \frac{1}{2} \\ b & 0 & \frac{1}{2} & 0 \end{bmatrix}, \tag{3.2a}$$

where $a + b = 1$. This matrix is not doubly stochastic (unless $a = \frac{1}{2}$), and the occupation probabilities are not equal at all, but (the labeling of states is as in fig. 2)

$$\boldsymbol{\pi} = \mathrm{col}\{\tfrac{1}{4}, \tfrac{1}{4}(a + \tfrac{1}{2}), \tfrac{1}{4}, \tfrac{1}{4}(b + \tfrac{1}{2})\}. \tag{3.2b}$$

Hence, although a doubly stochastic matrix is a *sufficient* condition for the validity of the MLBE, it is certainly not necessary.

To see what makes (1.3) true in the case of fig. 2, we note that we can write

$$\sum_{\beta} \langle x^2 \rangle_{\beta} \pi_{\beta} = \langle x^2 \rangle^* \pi^* + \sum_{\beta}{}' \langle x^2 \rangle_{\beta} \pi_{\beta}, \tag{3.3}$$

where the asterisk refers to the scattering site, and the primed summation runs only over regular sites. Since for all the regular sites $\langle x^2 \rangle_{\beta} = \frac{1}{2}$, we can write (3.3) as

$$\sum_{\beta} \langle x^2 \rangle_{\beta} \pi_{\beta} = \langle x^2 \rangle^* \pi^* + \tfrac{1}{2} \sum_{\beta}{}' \pi_{\beta} = \langle x^2 \rangle^* \pi^* + \tfrac{1}{2}(1 - \pi^*).$$

Doing the same for the denominator of (3.1), we find

$$\frac{D_x}{D_y} = \frac{\langle x^2 \rangle^* \pi^* + \frac{1}{2}(1 - \pi^*)}{\langle y^2 \rangle^* \pi^* + \frac{1}{2}(1 - \pi^*)} = \frac{a + \frac{1}{2}(1/\pi^* - 1)}{b + \frac{1}{2}(1/\pi^* - 1)},$$

which is identical to the result (1.4) of the MLBE if $1/\pi^* = 4$. This is indeed the case, as (3.2b) shows.

The MLBE is also valid for a two-dimensional lattice with $k \times k$ sites per cell, one of which is a scatterer (fig. 3). In this case $\pi^* = 1/k^2$ as the following argument shows[6]). The occupation probability π^* of the scatterer can be calculated as the limit (the scatterer is labeled as state 1)

$$\pi^* = \lim_{n \to \infty} p^{(2n)}(1 \mid 1), \tag{3.4}$$

where $p^{(2n)}(1 \mid 1)$ is the probability that the walker returned to the starting site while executing a random walk on the ILF of k^2 sites with periodic boundary conditions (return after an odd number of steps is impossible). We took the initial and final state to be identical since the limit (3.4) is independent of the initial state. Now consider the set S_n of all paths of $2n$ steps which lead the walker back to his starting site for the first time. We can split up the set S_n in a set $S_n^{(x)}$ consisting of all paths which start in the x-direction, and a set $S_n^{(y)}$ of paths starting in the y-direction. For every path w in $S_n^{(x)}$ there exists a path in $S_n^{(y)}$ which is obtained by reflecting the path w along the diagonal of the unit cell, and vice versa. Hence,the number of elements of the set $S_n^{(x)}$ equals that of $S_n^{(y)}$. Let us consider first the case that $a = b = \frac{1}{2}$. The probability of returning to the origin after $2n$ steps for the first time is

$$f^{(2n)}(1 \mid 1) = \left(\frac{1}{4}\right)^{2n} |S_n|, \tag{3.5}$$

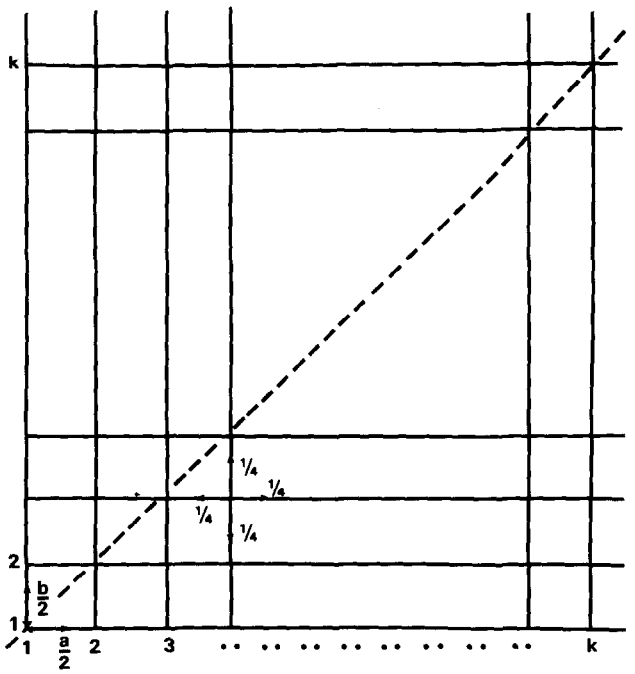

Fig. 3. ILF for a 2-D lattice with a square unit cell and one scatterer per ILF.

where $|S_n|$ is the number of elements in S_n. Now assume $a \neq b$. The probability $f^{(2n)}(1 \mid 1)$ can be written as a sum of paths which start in the x- and y-direction, respectively. Hence,

$$f^{(2n)}(1 \mid 1) = \frac{a}{2}\left(\frac{1}{4}\right)^{2n-1}|S_n^{(x)}| + \frac{b}{2}\left(\frac{1}{4}\right)^{2n-1}|S_n^{(y)}|$$

$$= \frac{1}{2}|S_n|\left(\frac{a}{2}+\frac{b}{2}\right)\left(\frac{1}{4}\right)^{2n-1} = \left(\frac{1}{4}\right)^{2n}|S_n|, \tag{3.6}$$

where we made use of the facts that $|S_n^{(x)}| = |S_n^{(y)}| = \frac{1}{2}|S_n|$, and $a + b = 1$. Hence, the result is the same as in the regular case (note that S_n does not depend on a). The same argument can be used for all walks which return to the origin, but not necessarily for the first time. So we see that $p^{(2n)}(1 \mid 1)$ is the same as in the regular case, hence also the limit π^*, and therefore

$$\pi^* = \frac{1}{k^2}, \tag{3.7}$$

since in the regular case all k^2 sites are equivalent. Eq. (3.7) is sufficient to guarantee the validity of the MLBE in this case, as used by Shuler and Mohanty[4]). The ratio of the corresponding diffusion coefficients is

$$\frac{D_y}{D_x} = \frac{b/2 + \frac{1}{4}(k^2-1)}{a/2 + \frac{1}{4}(k^2-1)} = 1 - \frac{2\phi}{\phi + \left(\frac{1+b/a}{1-b/a}\right)}, \tag{3.8}$$

where $\phi = 1/k^2$ is the concentration of scatterers [eq. (8) of ref. 5].

The argument leading to (3.7) can easily be extended to a d-dimensional lattice where the ILF is a hypercube of k^d sites, which contains one or more scattering sites (of the same type), *all lying on one of the main diagonals.* An example is the lattice of fig. 4a [3]). But in the other cases 4b,c the MLBE is invalid since the occupation probability π^* is affected by the anisotropy (see ref. 3, section 4).

There is a difference between the lattices of fig. 4a and figs. 4b,c in the sense that in the latter case there occur adjacent scatterers, i.e. there are direct scatterer–scatterer transitions. So one might wonder whether this is the reason for the breakdown of the MLBE. The answer turns out to be negative, as the example of fig. 5 shows. There are no adjacent scatterers on the lattice generated by the ILF of fig. 5. To test if the MLBE is valid, we only have to determine whether the occupation probability π^* of site 1 is equal to $\frac{1}{6}$. The matrix $\mathbf{T}$ for this case is

$$\mathbf{T} = \begin{bmatrix} 0 & \frac{1}{4} & \frac{1}{4} & \frac{1}{2} & 0 & 0 \\ \frac{1}{2}a & 0 & \frac{1}{4} & 0 & \frac{1}{2} & 0 \\ \frac{1}{2}a & \frac{1}{4} & 0 & 0 & 0 & \frac{1}{2} \\ b & 0 & 0 & 0 & \frac{1}{4} & \frac{1}{4} \\ 0 & \frac{1}{2} & 0 & \frac{1}{4} & 0 & \frac{1}{4} \\ 0 & 0 & \frac{1}{2} & \frac{1}{4} & \frac{1}{4} & 0 \end{bmatrix}$$

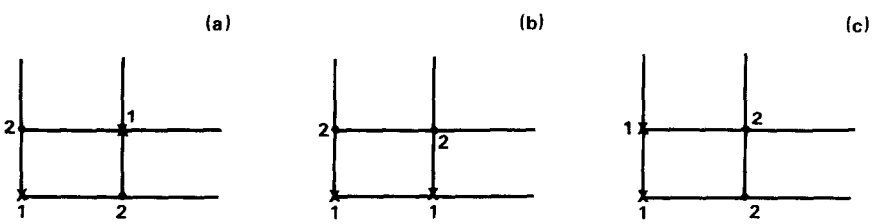

Fig. 4. ILF's for a lattice with anisotropic scatterers, all containing 2 scatterers per ILF.

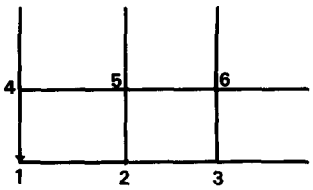

Fig. 5. Two-dimensional ILF with six sites and one scatterer.

and the vector π is given by

$$\pi = \pi^* \operatorname{col}\{1, c, c, d, e, e\},$$

where

$$c = 1 + \tfrac{6}{7}(a - \tfrac{1}{2}), \quad d = 1 - \tfrac{6}{7}(a - \tfrac{1}{2}), \quad e = 1 + \tfrac{2}{7}(a - \tfrac{1}{2})$$

and

$$\pi^* = [6 + \tfrac{5}{7}(1 - 2b)]^{-1}. \tag{3.9}$$

So π^* is not equal to $\frac{1}{6}$ (except in the completely regular case $a = b = \frac{1}{2}$) and the MLBE is not valid in this case. Moreover, there does exist an arrangement in an ILF of 6×6 sites with 6 scatterers on the diagonal for which MLBE is valid and therefore has different diffusion coefficients from the lattice of fig. 5, although the density is the same and although no adjacent scatterers occur. Hence, the conclusions drawn above remain valid, even if the arrangements with adjacent scatterers are excluded.

We conclude therefore that the MLBE is only valid in very special cases, which are all readily derived from the general results of ref. 2. Therefore, it is on the latter method that we have based our investigations of lattices with random arrangements of scatterers[3]).

Acknowledgements

The author thanks Prof. K.E. Shuler for many helpful discussions. This work was financially supported by the Netherlands Organization for the Advancement of Pure Research (Z.W.O.).

Appendix A

We prove here the result (1.9). Let

$$\Delta_{\nu_\beta}(i) = \begin{cases} 1, & \text{if the } i\text{th step of the walker is of type } \nu_\beta, \\ 0, & \text{otherwise}. \end{cases}$$

The number $n_{\nu_\beta}(n)$ of steps of type ν_β after a total of n steps is

$$n_{\nu_\beta}(n) = \sum_{i=1}^{n} \Delta_{\nu_\beta}(i),$$

so the average is

$$\langle n_{\nu_\beta}(n) \rangle = \sum_{i=1}^{n} \langle \Delta_{\nu_\beta}(i) \rangle . \tag{A.1}$$

Let $\langle \Delta_{\nu_\beta}(i) \mid \gamma \rangle$ be the conditional average of $\Delta_{\nu_\beta}(i)$, given that the ith transition occurs from internal state γ. Then we have

$$\langle \Delta_{\nu_\beta}(i) \mid \gamma \rangle = \delta_{\beta,\gamma} p_{\nu_\beta} , \tag{A.2}$$

independent of i. Here p_{ν_β} is the single-step probability of a transition of type ν_β. So, if $p_\gamma(i)$ is the probability that the walker is at a site with internal state γ just before the ith step, then

$$\langle n_{\nu_\beta}(n) \rangle = \sum_{i=1}^{n} \sum_{\gamma} \langle \Delta_\nu(i) \mid \gamma \rangle p_\gamma(i) = \sum_{i=1}^{n} p_{\nu_\beta} p_\beta(i) . \tag{A.3}$$

Now, suppose we let the initial distribution of the internal states be the equilibrium distribution, i.e. $p_\beta(0) = \pi_\beta$, where $\{\pi_\beta\}$ are the components of the eigenvector $\boldsymbol{\pi}$ of $\mathbf{T}$, as defined in section 1. Then $p_\beta(i) = \pi_\beta$, so from (A.3)

$$\langle n_\nu(n) \rangle = n p_{\nu_\beta} \pi_\beta . \tag{A.4}$$

If the initial distribution $p_\beta(0)$ is arbitrary, then (A.4) is true asymptotically as $n \to \infty$ as long as the embedded Markov chain on the set of internal states is *irreducible*, i.e. the matrix $\mathbf{T}$ in eq. (1.6) is irreducible [the argument is the same as that used in ref. 2].

Appendix B

In this appendix we give a proof of the lemma in section 2. Let there be m sites in a unit cell (or ILF, to be more precise). First we construct the transition matrix elements $T_{\alpha\beta}$. The diagonal elements $T_{\alpha\alpha}$ consist of two contributions. The first one is the probability t_α of a transition $(l, \alpha) \to (l, \alpha)$, i.e. a pause; secondly, there are a number, say $n_{\alpha\alpha}$, of transitions $(l, \alpha) \to (l', \alpha)$ where l and l' represent different unit cells, each having a probability $n_\alpha^{-1}(1 - t_\alpha)$ as stated in the conditions of the lemma. Thus

$$T_{\alpha\alpha} = \frac{n_{\alpha\alpha}}{n_\alpha}(1 - t_\alpha) + t_\alpha . \tag{B.1a}$$

The off-diagonal elements $T_{\alpha\beta}$ consist of all the transitions, say $n_{\alpha\beta}$ in number, from a site (l, β) to (l', α) with $\alpha \neq \beta$, but where l and l' may be identical. Each of these transitions also has a probability $n_\beta^{-1}(1 - t_\beta)$. Thus

$$T_{\alpha\beta} = \frac{n_{\alpha\beta}}{n_\beta}(1 - t_\beta) \quad (\alpha \neq \beta) . \tag{B.1b}$$

Consistency requires that the total number n_β of transitions with nonzero displacement satisfies the equality

$$n_\beta = \sum_\alpha n_{\alpha\beta} . \tag{B.2}$$

The assertion is now that the occupation probabilities $\{\pi_\beta\}$ are given by (2.6). That is, we have to check that the vector $\boldsymbol{\pi}$ is a right eigenvector of the matrix $\mathbf{T}$ with matrix elements (B.1) corresponding to the eigenvalue $\lambda_0 = 1$. The proof is straightforward:

$$\begin{aligned}
\sum_\beta T_{\alpha\beta}\pi_\beta &= \sum_\beta \left\{\frac{n_{\alpha\beta}}{n_\beta}(1 - t_\beta) + \delta_{\alpha\beta}t_\alpha\right\} c n_\beta (1 - t_\beta)^{-1} \\
&= c\left\{\sum_\beta n_{\alpha\beta} + t_\alpha n_\alpha (1 - t_\alpha)^{-1}\right\} \\
&= c\left\{\sum_\beta n_{\beta\alpha} + t_\alpha n_\alpha (1 - t_\alpha)^{-1}\right\} \\
&= c\{n_\alpha + t_\alpha n_\alpha (1 - t_\alpha)^{-1}\} = c n_\alpha (1 - t_\alpha)^{-1} = \pi_\alpha .
\end{aligned} \tag{B.3}$$

Here we have used that $n_{\alpha\beta} = n_{\beta\alpha}$ in agreement with the requirement that for every transition between different sites also the reverse transition is allowed. The constant in (2.6) is fixed by normalization of the occupation probabilities $\{\pi_\beta\}$. This finishes the proof.

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6) The result also follows from eq. (4.9) of ref. 3 since $\tilde{K}(a') = 0$ for $a' = \frac{1}{4}$ as is the case here.