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Asymptotic Properties of Multistate Random Walks. II. Applications to Inhomogeneous Periodic and Random Lattices

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The previously developed formalism for the calculation of asymptotic properties of multistate random walks is used to study random walks on several inhomogeneous periodic lattices, where the periodically repeated unit cell contains a number of inequivalent sites, as well as on lattices with a random distribution of inequivalent sites. We concentrate on the question whether the random walk properties depend on the spatial arrangement of the sites in the unit cell, or only on the number density of the different types of sites. Specifically we consider lattices with periodic and random arrangements of columns and lattices with periodic and random arrangements of anisotropic scatterers.

KEY WORDS: Multistate random walks; inhomogeneous periodic and random lattices; random jump rates.

1. INTRODUCTION

In a previous paper, hereafter referred to as I,[1] we have investigated the asymptotic behavior of various random walk properties for multistate random walks on inhomogeneous periodic lattices. These random walks are characterized by the fact that the walker can be in a number of internal states. Our main interest has been the case of nonequivalent *configurational* internal states (i.e., sites) within a periodically repeated unit cell. In I expressions for the asymptotic mean and (co)variance of the displacement of the walk have been derived, both in discrete and continuous time. The results for the (co)variances simplify considerably for walks which are

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locally unbiased in every direction, i.e., walks in which the average single-step displacements from every site in every space direction are zero.[3] In addition, the long-time behavior of the probability of return to the origin and the expected number of distinct sites visited has been obtained. The time dependence of these quantities for inhomogeneous periodic lattices is the same as for perfect periodic lattices and only the coefficients are modified.

In this paper we consider in detail the question which was raised in the introduction of I, namely, the condition(s) under which the asymptotic random walk properties do, or do not, depend on the spatial arrangement of the sites within a unit cell.[4] In view of the results of I, we limit ourselves to a restricted number of properties (mainly the mean square displacements) since other properties, such as the probability of return to the origin or the expected number of distinct sites visited, are determined by the diffusion coefficients associated with the mean square displacements. To gain some insight into the problem, we study here a number of representative examples.

In Section 2, we discuss the case of sparsely periodic two-dimensional lattices and its extension to the case of a random distribution of columns. Also, a modified sparsely periodic lattice, the "brick lattice," is studied, which is obtained from a sparsely periodic lattice by shifting some of the vertical bonds a fixed number of sites along the horizontal direction. For the latter case we show that the random walk properties are no longer independent of the detailed spatial arrangement of the shifted bonds, in contrast to the original sparsely periodic lattice, where only the *density* of vertical columns is relevant. In Section 3 we consider the problem of lattices with anisotropic scatterers, both for periodic and random arrangements. Some analytical results for the case of low or high density of scatterers are presented. Section 4 contains a summary and discussion of the results obtained in this paper and a comparison with earlier results in the literature.

2. SPARSELY PERIODIC LATTICES

2.1. Periodic Distribution of Columns

In Section 2.2 of I we discussed an example of a so-called sparsely periodic lattice in two dimensions. As can be seen from Fig. 1, a number of vertical columns is deleted in a periodic way so that the walker can only step in the y direction on a subset of sites.

[3] That is, the walk has the Martingale property.[2]

[4] We will be concerned here only with random walks having a zero drift velocity in every space direction.

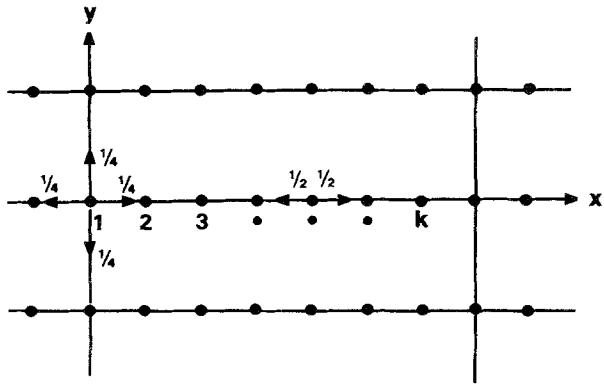

Fig. 1. Sparsely periodic lattice with horizontal periodicity k; step probabilities are as indicated.

Various random walk properties on this inhomogeneous periodic lattice, i.e., mean, variances, occupation probabilities, expected number of distinct sites visited, probability of return to the origin, were worked out in the long-time limit in previous papers.[1,3,5] One of us postulated in an earlier paper[3] that these results should be independent of the arrangement of the vertical columns for a fixed number density of columns. This was later confirmed in Ref. 5 and rigorously proved to be true in Refs. 6–8.

We now want to demonstrate that the methodology developed by us in our preceding paper[1] yields, in a very simple manner, the same results, while at the same time giving more insight into the underlying reasons for the lack of dependence of random walk properties on the detailed spatial arrangement of columns.

As a first step in this demonstration, we consider a generalization of the lattice in Fig. 1, where the unit cell now contains k columns which all differ from each other, i.e., from a site on the βth column ($\beta = 1, 2, \ldots, k$) the walker jumps with a probability p_β in the positive or negative x direction, and with probability $q_\beta = \frac{1}{2} - p_\beta$ in the positive or negative y direction. The unit cell or, to be more precise, the irreducible lattice fragment (ILF), for this lattice is shown in Fig. 2. To determine the mean square displacements,

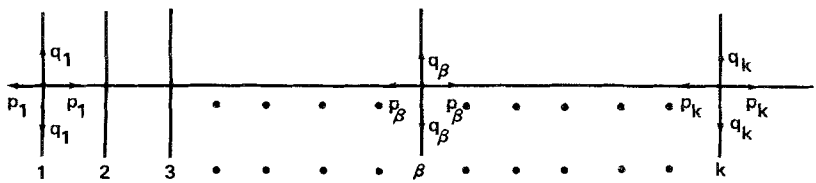

Fig. 2. ILF for a periodic lattice with column-dependent step probabilities; horizontal periodicity k.

we first construct the matrix T, which describes the transitions between the internal states, and calculate[1] its right eigenvector π corresponding to the eigenvalue $\lambda_0 = 1$. We find

$$\mathsf{T} = \begin{pmatrix} 2q_1 & p_2 & & & p_k \\ p_1 & 2q_2 & \ddots & & \\ & p_2 & \ddots & \ddots & \\ & & \ddots & \ddots & p_k \\ p_1 & & p_{k-1} & & 2q_k \end{pmatrix}, \qquad \pi = c \begin{pmatrix} p_1^{-1} \\ p_2^{-1} \\ \cdot \\ \cdot \\ \cdot \\ p_k^{-1} \end{pmatrix} \tag{2.1}$$

where the constant c is determined by normalization:

$$c = \left(\sum_{\beta=1}^{k} p_\beta^{-1} \right)^{-1} \tag{2.2}$$

One can easily check that the vector π in (2.1) is indeed the right eigenvector of T corresponding to the eigenvalue $\lambda_0 = 1$ (note that $2p_\beta + 2q_\beta = 1$, all β). The simple form of the equilibrium occupation probabilities $\{\pi_\beta\}$ is due to the validity of *detailed balance* for this model, i.e., $p_1\pi_1 = p_2\pi_2 = \cdots = p_k\pi_k$, which obviously does not involve the transitions in the y direction, since these constitute jumps from an internal state to itself.

To find the diffusion coefficients, we first calculate the one-step variances from each state β. These are easily found to be

$$\langle x^2 \rangle_\beta = 2p_\beta, \qquad \langle y^2 \rangle_\beta = 2q_\beta, \qquad \langle xy \rangle_\beta = \langle yx \rangle_\beta = 0 \tag{2.3}$$

where we have set the lattice constants equal to unity. Then by applying Eq. (2.2.31) of I (the walk is locally unbiased), we have

$$D_{xx} = \tfrac{1}{2} \sum_\beta \langle x^2 \rangle_\beta \pi_\beta = ck \tag{2.4a}$$

$$D_{yy} = \tfrac{1}{2} \sum_\beta \langle y^2 \rangle_\beta \pi_\beta = c \sum_{\beta=1}^{k} \frac{q_\beta}{p_\beta} = \tfrac{1}{2} - D_{xx} \tag{2.4b}$$

$$D_{xy} = D_{yx} = 0 \tag{2.4c}$$

in agreement with Corollary 1 of Ref. 6, and[5]

$$\frac{\langle x^2(n) \rangle}{\langle y^2(n) \rangle} \sim \frac{D_{xx}}{D_{yy}} = \frac{k}{\sum_\beta (q_\beta / p_\beta)} \qquad (n \to \infty) \tag{2.5a}$$

[5] The relevance of these results for Shuler's bond enumeration method[3] is discussed in Ref. 9.

For the sparsely periodic lattice of Fig. 1, $q_1 = p_1 = \frac{1}{4}$ and $q_2 = \cdots = q_k = 0$, so that

$$\frac{\langle x^2(n)\rangle}{\langle y^2(n)\rangle} \sim k \qquad (n \to \infty) \tag{2.5b}$$

as already found in I. It can readily be seen from (2.4) and (2.5a) that the variances and the diffusion coefficients are independent of the spatial arrangement of the columns. If a waiting time density with finite mean waiting time is assigned to each of the sites in Fig. 2, the ratio of asymptotic mean square displacements in the x and y direction is still given by the right-hand side of (2.5), as can be deduced from Eq. (3.2.9a) of I.

The probability of return to the origin after n steps, denoted now by $R_n(k)$ to indicate the dependence on the number k of columns per unit cell, averaged over initial states, is given by Eq. (2.3.11) of I as

$$R_n(k) \overset{n\to\infty}{\sim} k^{-1}(\det 2\mathsf{D})^{-1/2}(\det \mathsf{A})(2\pi n)^{-1} \tag{2.6}$$

Here D is the diffusion matrix and A has matrix elements

$$A_{ij} = \mathbf{a}_i \cdot \mathbf{e}_j \tag{2.7}$$

where $\{\mathbf{a}_i\}$ is the set of fundamental translation vectors which define the unit cell and $\{\mathbf{e}_i\}$ is the set of basis vectors which is used to define the coordinate system.[1] Actually (2.6) should be multiplied by a factor $\{1 + (-)^n\}$, but since we are concerned here with ratios for the same value of n, this factor is unimportant. From (2.6) we find for the ILF of Fig. 2,

$$h(k) \equiv \frac{R_n(k)}{R_n(1)} \overset{n\to\infty}{\sim} k^{-1}\left[\frac{\det 2\mathsf{D}(k)}{\det 2\mathsf{D}(1)}\right]^{-1/2} \frac{\det \mathsf{A}(k)}{\det \mathsf{A}(1)} \tag{2.8}$$

The elements of the matrix $\mathsf{A}(k)$, as defined in (2.7), are

$$A_{xx}(k) = k, \qquad A_{yy}(k) = 1, \qquad A_{xy} = A_{yx} = 0$$

so that

$$\det \mathsf{A}(k) = k \tag{2.9}$$

Since the diffusion matrix is diagonal, we finally obtain

$$h(k) = \left[\frac{D_{xx}(k)\, D_{yy}(k)}{D_{xx}(1)\, D_{yy}(1)}\right]^{-1/2} \tag{2.10}$$

This result confirms the *assumption* made in Ref. 3 that the probability of return to the origin is inversely proportional to the area $[\langle x^2(n)\rangle\langle y^2(n)\rangle]^{1/2}$ covered by the random walker. However, as can be seen from (2.8), the simple result (2.10) will not necessarily hold for other types of lattices, since the diffusion matrix is not necessarily diagonal, nor is the factor $\det \mathsf{A}(k)/k \det \mathsf{A}(1)$ always equal to unity. A similar remark applies to the expected number of distinct sites visited (summed over final and averaged over initial internal states), which is in this case proportional to the area $[\langle x^2(n)\rangle\langle y^2(n)\rangle]^{1/2}$, as assumed in Eq. (15) of Ref. 3. However, in view of Eq. (2.4.10b) of I which involves the complete generating function $G_{\alpha\alpha}(\mathbf{0}, 1)$ and not merely the diffusion matrix, an analogous result in three dimensions is not to be expected, contrary to the remarks of Ref. 3, Eq. (51).

We note that the asymptotic results for the probability of return to the origin and the expected number of distinct sites visited are independent of the arrangement of columns since they are functions only of the diffusion coefficients for which the arrangement has already been shown to be irrelevant.

2.2. Random Distribution of Columns

We now turn our attention to the explicit calculation of the diffusion coefficients for lattices of the type shown in Fig. 2 with a *random* distribution of columns. Let us consider a two-dimensional lattice with two types of columns, denoted by 1 and 2. For columns of type 1, the stepping probabilities in the x and y direction are p_1 and q_1, respectively, and p_2, q_2 for columns of type 2.

The basic approach is as follows. We start by assuming a random distribution of the columns in a unit cell with k sites, which is then periodically repeated as before. The diffusion coefficients, which can be calculated for every realization of this random distribution by using Eq. (2.4) above, are now random variables. We will show that in the limit of an infinite number of columns per unit cell (i.e., $k \to \infty$), the diffusion coefficients approach a nonrandom limit, which is identical for almost all realizations of the random distribution of columns.

Let us assume that we have an inhomogeneous periodic lattice of k sites per unit cell as in Fig. 2, where now the $\{p_\beta\}$ and $\{q_\beta\}$ are random, i.e., for any $\beta = 1, 2, \ldots, k$, $p_\beta = p_1$, $q_\beta = q_1$ with probability ν and $p_\beta = p_2$, $q_\beta = q_2$ with probability $1 - \nu$, independent of the values of the other p's and q's (i.e., no correlations between columns). In an inhomogeneous periodic lattice with a *fixed* number s_1 of columns of type 1 and a fixed

number $k-s_1$ of columns of type 2 in each unit cell, the diffusion coefficients have the form

$$D_x(s_1)=k\left[\left(\frac{s_1}{p_1}+\frac{k-s_1}{p_2}\right)\right]^{-1}=\left[\frac{s_1}{k}\cdot\frac{1}{p_1}+\left(1-\frac{s_1}{k}\right)\frac{1}{p_2}\right]^{-1} \tag{2.11a}$$

$$D_y(s_1)=\frac{1}{2}-D_x(s_1) \tag{2.11b}$$

For the present case of a random distribution of columns in the unit cells of an inhomogeneous periodic lattice, the formulas (2.11) still hold with the understanding that the number s_1 is now a *random variable*, taking values 0, 1,..., k, and distributed according to a binomial distribution. We now let the size of the unit cell increase without bound by taking the limit $k\to\infty$ with v fixed. By the Strong law of large numbers the fraction s_1/k of columns of type 1 then approaches the (nonrandom) limit v for almost all realizations of the random distribution of columns. Thus with probability 1,

$$D_x \xrightarrow{k\to\infty} [vp_1^{-1}+(1-v)\,p_2^{-1}]^{-1} \tag{2.12}$$

$$D_y \xrightarrow{k\to\infty} \tfrac{1}{2}-[vp_1^{-1}+(1-v)\,p_2^{-1}]^{-1} \tag{2.13}$$

Comparing (2.11) and (2.12), we see that the diffusion coefficients for the random distribution of columns are identical to those for a periodic arrangement of columns, provided that the *fractions* s_1/k and s_2/k for the *periodic* case are equal to the *probabilities* v and $(1-v)$ for the random case, where we take v to be a rational number.

The only question which remains at this point is whether the above procedure of starting with a periodic random arrangement with a subsequent passage to the limit of an infinite period, i.e., $k\to\infty$, leads to the correct result for the infinite random system. As has been shown by den Hollander,[10] the answer is "yes" for almost all realizations of the random distribution of columns; in addition, he found that the relations between the diffusion coefficients and the probability of return to the origin or the expected number of distinct sites visited, as established for periodic inhomogeneous lattices in I, are still valid for the lattice with a random arrangement of columns.

A result of a more general nature was recently derived by Vigfusson.[11] He showed that under fairly mild conditions the *expectation value* $E\{D^{(m)}\}$ (where the expectation is over the random distribution of inequivalent sites or jump rates) of the diffusion constant $D^{(m)}$ for a one-dimensional chain with periodicity m approaches the expectation value

$E\{D^{(\infty)}\}$ of the infinite chain as $m \to \infty$. A similar proof can be given for higher dimensional lattices.[12] In the examples of this section and of Section 3, we have the much stronger result that almost all realizations of the infinite random system have the same diffusion coefficients, but this is not always the case.[13,15,25,26]

2.3. Modified Sparsely Periodic Lattice (The Brick Lattice)

It is of some interest to discuss what happens if, instead of deleting whole columns from a square lattice, one deletes individual vertical bonds, thus obtaining a type of percolation lattice. As a first step, we consider here the following rudimentary percolation lattice: start from a sparsely periodic lattice (horizontal periodicity k) and move the vertical bonds $(m-1)$ sites to the right in every other horizontal strip, with $1 \leqslant m \leqslant k+1$ (Fig. 3). In this way, the density of vertical bonds is unaltered and the question is whether the random walk properties on this lattice, which we denote as the „brick lattice," are affected by this shift.

The random walk is defined by the following single-step transition probabilities: The walker jumps with probability 1/2 to each of his two neighbors from a site without vertical bonds, and with probability 1/3 to each of his three neighbors from a site with a vertical bond. It can easily be

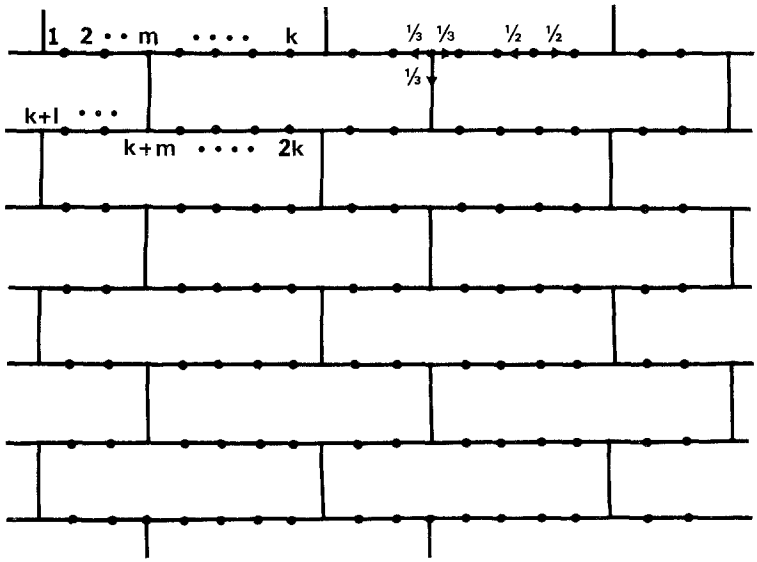

Fig. 3. Brick lattice, horizontal periodicity $k_x = k$, vertical periodicity $k_y = 2$. Step probabilities are as indicated.

shown that under these assumptions the occupation probabilities of the $2k$ sites in the unit cell are (see Ref. 9)

$$\pi_\beta^{(2)} = 2/c' \tag{2.14a}$$

for sites with two bonds connected to it, and

$$\pi_\beta^{(3)} = 3/c' \tag{2.14b}$$

for sites with three bonds, where from normalization

$$c' = (4)(3) + (2k-4)2 = 4(k+1) \tag{2.14c}$$

Since the walk is locally unbiased in the horizontal direction, i.e., $\langle x \rangle_\beta = 0$ for all β, we immediately have from Eq. (2.2.31) of I

$$\begin{aligned} 2D_x &= \sum_\beta \langle x^2 \rangle_\beta \pi_\beta = (4)\left(\frac{2}{3}\right)\left(\frac{3}{c'}\right) + (2k-4)(1)\left(\frac{2}{c'}\right) \\ &= \frac{4k}{c'} = \frac{k}{k+1} \end{aligned} \tag{2.15}$$

Hence the diffusion coefficient in the horizontal direction is the same as for the original sparsely periodic lattice, and independent of m.

The diffusion coefficient D_y in the vertical direction is more difficult to obtain owing to the fact that the walk is not locally unbiased in the vertical direction. A formula has been derived in I [Eq. (2.2.29b)] for this case, which involves all the eigenvalues and eigenvectors of the $2k$-dimensional transition matrix, but these are hard to calculate. Instead we use the following stratagem to circumvent this problem. The diffusion matrix D can be expressed as [Eq. (2.2.24) of I with $m_k = 0$]

$$2\mathsf{D} = \mathsf{A}^T \mathsf{S} \mathsf{A} \tag{2.16}$$

where the matrix A (with transpose A^T) is defined in (2.7) and the matrix S depends only on the connectivity of the lattice [see Eq. (2.2.13b) of I]. We now deform the lattice of Fig. 3 in such a way as to give rise to a locally unbiased random walk while preserving the original connectivity (Fig. 4).

For the deformed lattice we have

$$2\mathsf{D}' = \mathsf{A}'^T \mathsf{S} \mathsf{A}' \tag{2.17}$$

where S is the same as in (2.16) and all primed quantities pertain to the deformed lattice. Hence from (2.16) and (2.17)

$$2\mathsf{D} = \mathsf{A}^T (\mathsf{A}'^T)^{-1} (2\mathsf{D}') (\mathsf{A}')^{-1} \mathsf{A} \tag{2.18}$$

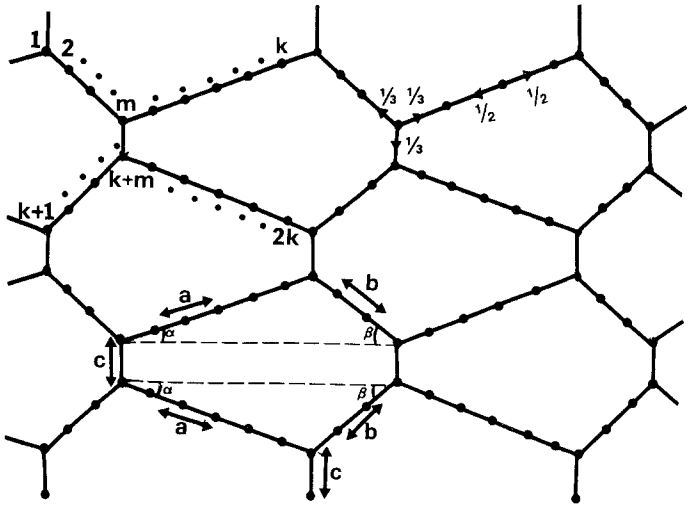

Fig. 4. Deformed lattice corresponding to Fig. 3. The ILF consists of the $2k$ labeled sites.

Since the deformed lattice is by construction locally unbiased in all directions, we can apply Eq. (2.2.31) of I, involving only the eigenvector π' of the transition matrix T′ corresponding to the deformed lattice, to calculate D′. In this way we can obtain D as well, since the matrices A and A′ are determined completely by the geometry of the lattice. The details are presented in Appendix A. The result is

$$2D_x = \frac{k}{k+1} \tag{2.19}$$

as we already found, and

$$2D_y = \frac{1}{k+1} \cdot \frac{1}{\gamma} \tag{2.20a}$$

where

$$\gamma = k^{-1}[km - (m-1)^2] \tag{2.20b}$$

We thus see that D_y *does depend on* m, i.e., the spatial arrangement of the vertical bonds. The sparsely periodic case is recovered by putting $m = 1$ or $k+1$ for which $\gamma = 1$. In Fig. 5 we plot the ratio $D_x/D_y = k\gamma$ as a function of m. This ratio attains its maximum when $m = k/2 + 1$ (if k is even), corresponding to the case of largest anisotropy in the diffusion.

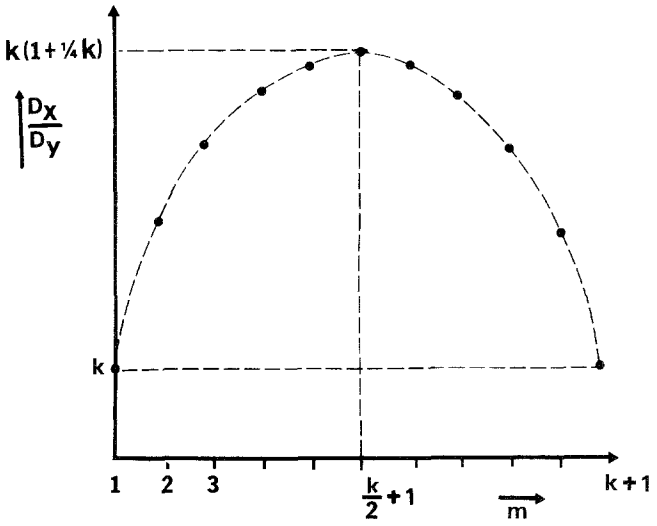

Fig. 5. Ratio of the diffusion coefficients in the horizontal and vertical direction as a function of the position m of shifted bonds.

The first conclusion which can be drawn is that it makes a considerable difference whether one shifts vertical columns as a whole, as in the previous sections, or individual vertical bonds, as in the brick lattice. Secondly, our initial question whether the asymptotic random walk properties do or do not depend on the arrangement of the bonds has to be refined somewhat. It turns out, as in the example just discussed, that some of these properties may depend upon arrangement while others do not. In the case of the brick lattice, D_y does depend on the arrangement of shifted bonds, but D_x does not. The occupation probabilities (2.14) are independent of arrangement. From the results of Ref. 9 it follows that the average numbers $\langle n_x(n)\rangle$ and $\langle n_y(n)\rangle$ of steps in the x and y direction after n steps are for the case of the brick lattice given asymptotically by

$$\langle n_x(n)\rangle \sim \left(\frac{k}{k+1}\right) n, \qquad \langle n_y(n)\rangle \sim \left(\frac{1}{k+1}\right) n \qquad (n \to \infty) \quad (2.21)$$

a result which is also independent of arrangement. Since the walk is locally unbiased in the x direction, the horizontal variance $\langle x^2(n)\rangle$ is directly proportional to $\langle n_x(n)\rangle$, which immediately leads to (2.15). However, since the walk is not locally unbiased in the y direction, the lack of dependence of $\langle n_y(n)\rangle$ on arrangement of shifted bonds does not imply a similar independence for D_y. We have not yet addressed the more difficult problem of obtaining D_y for brick lattices with randomly distributed vertical bonds.

3. LATTICES WITH ANISOTROPIC SCATTERERS

3.1. Periodic Arrangements of Scatterers

In this section we consider lattices with anisotropic scatterers, a topic already discussed by Shuler and Mohanty for a special case.[4] The main difference from the sparsely periodic lattices of Section 2.1 turns out to be that the arrangement of the scatterers in the unit cell now matters, i.e., inhomogeneous periodic lattices with the same density of scatterers but different arrangement may have different diffusion coefficients. This will be illustrated by a number of examples. In the subsequent sections, we discuss the case of a random arrangement of scatterers. This is much more difficult to deal with than the random arrangement of columns discussed in Section 2.2, and we limit ourselves, for now, to some general qualitative remarks as well as some low-density results.

Let us start by considering a two-dimensional lattice with four sites per unit cell, two of which are scatterers. One of the possible lattices is given in Fig. 6. The transition probabilities from a scatterer are a in the horizontal and b in the vertical directions, where $2a + 2b = 1$. The ILF for this lattice is given in Fig. 7a. In Figs. 7b and 7c, the other two possible ILF's with four sites and two scatterers are drawn. The latter two ILF's transform into each other by a simultaneous interchange $x \leftrightarrow y$, $a \leftrightarrow b$. This manifests itself in the results (3.2) for the occupation probabilities. One of the scatterers can always be placed at the origin without loss of generality, and therefore three possibilities for the second scatterer remain.

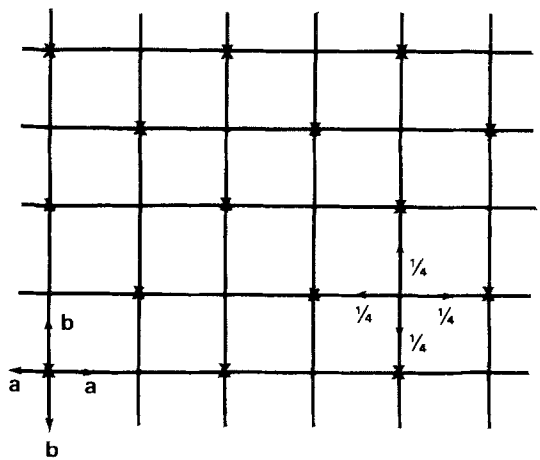

Fig. 6. Two-dimensional anisotropic lattice. x: scattering sites. The jump probabilities are indicated.

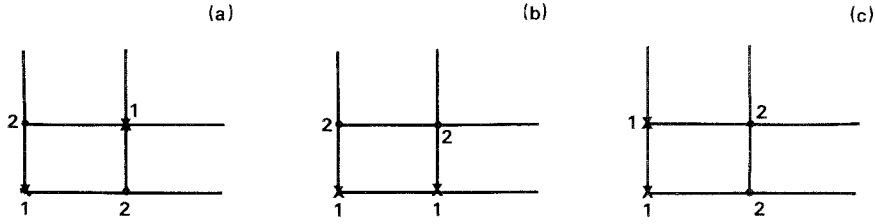

Fig. 7. ILF's with four sites and two scatterers in three different arrangements, (a)–(c). Jump probabilities for crosses and circles are as in Fig. 6.

The diffusion coefficients in all three cases can be written as[1]

$$D_x \equiv \tfrac{1}{2}\sum_{\beta} \langle x^2 \rangle_\beta \, \pi_\beta = \tfrac{1}{2}[2a\pi_1 + \tfrac{1}{2}(1-\pi_1)] \tag{3.1a}$$

$$D_y = \tfrac{1}{2} - D_x \tag{3.1b}$$

where π_1 is the occupation probability of site 1 (the scatterer) in Figs. 7a–c, and $\pi_2 = 1 - \pi_1$, that of site 2 (the regular site). Note that we have reduced the ILF of four sites to an ILF of two sites in all three cases, and used that $\langle x^2 \rangle_1 = 2a$, $\langle x^2 \rangle_2 = \frac{1}{2}$. So we need only to determine the occupation probability π_1 of the scatterer in all three cases, i.e., calculate the eigenvector $\boldsymbol{\pi}$ of the matrix T which describes the transitions between the internal states. For the three cases of Fig. 7, we find the following:

$$\text{Fig. 7a,} \quad \mathsf{T} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \boldsymbol{\pi} = \frac{1}{2}\begin{pmatrix} 1 \\ 1 \end{pmatrix} \tag{3.2a}$$

$$\text{Fig. 7b,} \quad \mathsf{T} = \begin{pmatrix} 2a & \frac{1}{2} \\ 2b & \frac{1}{2} \end{pmatrix}, \quad \boldsymbol{\pi} = \frac{1}{1+4b}\begin{pmatrix} 1 \\ 4b \end{pmatrix} \tag{3.2b}$$

$$\text{Fig. 7c,} \quad \mathsf{T} = \begin{pmatrix} 2b & \frac{1}{2} \\ 2a & \frac{1}{2} \end{pmatrix}, \quad \boldsymbol{\pi} = \frac{1}{1+4a}\begin{pmatrix} 1 \\ 4a \end{pmatrix} \tag{3.2c}$$

We see that the occupation probability π_1 is *different* for all three lattices of Fig. 7. Similar results can be shown to hold for arrangements of two scatterers in a unit cell of 3×3 sites, etc. *Thus the diffusion coefficients and the other random walk properties discussed in I differ for different periodic arrangements of scatterers with identical density.* It is evident that this remark is general and not just limited to the specific examples discussed above. It is therefore not possible to apply the simple method of Section 2.2 to the case of the random distribution of anisotropic scatterers at a fixed average density. Some low-density results are presented in Section 3.3.

The consequences of these results for the validity of the modified bond enumeration method of Shuler and Mohanty[4] are discussed in Ref. 9.

3.2. Random Arrangement of Scatterers

The results presented above raise the following important question. It is a well-known experimental observation, and a necessary condition for the existence of modern technology based on solid state materials, that macroscopic properties, such as transport coefficients, for solids with defects (scatterers, impurities, holes, etc.) at a given density are characteristic properties of the material and do not depend upon where or how the material was prepared. For instance, amorphous semiconductors of a given composition with, say, 10% recombination centers randomly distributed in the material, will yield the same results for the photocurrent in different laboratories, even though the material may have been prepared by different techniques. This appears to be in conflict with the results presented above for periodic arrangements of scatterers.

For anisotropic scatterers, the answer to this apparent dichotomy can be inferred from the results of Lawler[14] on the convergence of a random walk in a random environment of anisotropic scatterers. Before discussing his result, we first note that the diffusion coefficient in the x direction in any of the above examples of a periodic arrangement of anisotropic scatterers with arbitrary density ρ, $0 < \rho < 1$, per unit cell, can be written[1]

$$D_x \equiv \tfrac{1}{2} \sum_{\beta} \langle x^2 \rangle_\beta \, \pi_\beta = \tfrac{1}{2} \langle x^2 \rangle^* \, \pi^*(m, \rho) + \tfrac{1}{2} \langle x^2 \rangle \, \pi(m, \rho) \tag{3.3}$$

where m denotes the total number of sites in the unit cell and where $\langle x^2 \rangle^* = 2a$, $\langle x^2 \rangle = \frac{1}{2}$. The occupation probabilities $\pi^*(m, \rho)$ and $\pi(m, \rho)$ are given by

$$\pi^*(m, \rho) = \sum_{\beta}^{*} \pi_\beta, \qquad \pi(m, \rho) = 1 - \sum_{\beta}^{*} \pi_\beta \tag{3.4}$$

where the asterisk indicates a summation over all the *scattering* sites in the unit cell. In other words, $\pi^*(m, \rho)$ and $\pi(m, \rho)$ are the asymptotic probabilities that the walker is, respectively, on a scattering or a regular site.

The work of Lawler shows that if one considers a random walk on a lattice with a stationary and ergodic distribution μ of anisotropic scatterers, the occupation probabilities of the scatterers and regular sites are, *for almost all realizations of* μ, equal to constants $\bar{\pi}^*$ and $1 - \bar{\pi}^*$, respectively, where $\bar{\pi}^*$ depends on μ. A special case of a stationary ergodic distribution is the *random distribution* $\mu^{(R)}$, where the probability that a site is a scatterer or regular site is given by ν and $1 - \nu$, respectively, independent of the other sites.

From Lawler's result and our findings in Section 3.1, we draw two conclusions:

(i) Different realizations of a *random* distribution $\mu^{(R)}$ of scatterers with identical density v of scatterers give rise to identical diffusion coefficients with the exception of a set of measure zero.

(ii) The periodic arrangements with density v of scatterers, being possible realizations of the random distribution $\mu^{(R)}$, belong to this set of measure zero.[6]

There is thus no contradiction between the experimental observation of the well-characterized macroscopic (i.e., averaged) properties of defect materials and the theoretical results on inhomogeneous periodic lattices with anisotropic scatterers as given above. Unless very special techniques are employed for the specific purpose of creating a material with periodic arrangements of "defects," the preparation of such material will lead, for almost all realizations, to a random arrangement of defects.

A slightly weaker form of Lawler's result has recently been proven for the case where some of the transition probabilities are zero, i.e., the percolation problem, where it has been shown that the diffusion coefficient for a random walk starting on the infinite cluster, i.e., above the percolation threshold, is independent (in probability) of the configuration of the broken bonds.[15] A similar result is to be expected in the case of the brick lattice of Section 2.3, when the vertical bonds are shifted *randomly* in the horizontal direction.

Returning to the lattice with a random distribution of anisotropic scatterers, we see that in order to calculate the diffusion coefficients we need to calculate the occupation probability $\bar{\pi}^*(v)$ of scatterers. This is a very difficult problem for arbitrary v, $0 \leqslant v \leqslant 1$. We study some limiting cases in the next section.

3.3. Low-Density Results

In order to calculate the occupation probability $\bar{\pi}^*(v)$ of the scatterers in the case of a random distribution of scatterers, we assume that v is very close to 0 or 1. Then we can use perturbation theory to obtain an analytical expression for $\bar{\pi}^*(v)$. The calculation is outlined in Appendix B for the slightly more general case where the step probabilities from a "scatterer" are, respectively, a and b in the horizontal and vertical directions, as before, but where from the "regular" sites the step probabilities are, respectively, a' and b' in the horizontal and vertical directions. (The case dis-

[6] Strictly speaking, there is the remote possibility that some periodic arrangements might have the same diffusion coefficients as in the random case.

cussed in Sections 3.1 and 3.2 is recovered by putting $a' = b' = \frac{1}{4}$.) The result is[7]

$$\bar{\pi}^*(v) = v[1 + (a - a')\, K(a')]^{-1} + O(v^2) \qquad (v \to 0) \tag{3.5}$$

where

$$K(a') = \frac{2}{\pi^2} \int_0^\pi d\theta_1 \int_0^\pi d\theta_2 \left[\frac{(1 - \cos\theta_1) - (1 - \cos\theta_2)}{2a'(1 - \cos\theta_1) + (1 - 2a')(1 - \cos\theta_2)} \right]$$

$$= 4 \left[\frac{1}{2a'\pi} \arctan\left(\frac{2a'}{1 - 2a'} \right)^{1/2} - \frac{1}{(1 - 2a')\pi} \arctan\left(\frac{1 - 2a'}{2a'} \right)^{1/2} \right] \tag{3.6}$$

Thus the asymptotic probability $\bar{\pi}^*(v)$ that the walker is on a scattering site is, even for small v, in general different from the probability v that a site is a scatterer. An exception is the case $a' = \frac{1}{4} = b'$ considered by Shuler and Mohanty,[4] since $K(\frac{1}{4}) = 0$. The result for v close to 1 can be obtained from (3.5) by replacing v by $1 - v$ and interchanging a and a'. Higher-order corrections to (3.5) can in principle be calculated from the result (B30) of Appendix B by cluster expansion methods of statistical mechanics. These corrections involve *self-correlations*, corresponding to repeated visits of the walker to the same internal state (site), and are therefore of order v^2, v^3, etc.

Let us now compare these results to the situation of a *periodic* arrangement with one scatterer in a square unit cell of m^2 sites. The occupation probability of the scatterer is obtained in Appendix B as

$$\pi^*(\rho) = \rho[1 + (a - a')\, \tilde{K}(a')]^{-1} \tag{3.7}$$

where $\rho = 1/m^2$ is the density of scatterers, and

$$\tilde{K}(a') = \frac{1}{m^2} \sum_{k,l}{}' \left\{ \frac{[1 - \cos(2\pi k/m)] - [1 - \cos(2\pi l/m)]}{2a'[1 - \cos(2\pi k/m)] + (1 - 2a')[1 - \cos(2\pi l/m)]} \right\} \tag{3.8}$$

where $k, l = 0, 1, 2, \ldots, m - 1$, and the prime indicates that the term with $k = l = 0$ is omitted. For large m, i.e., for $\rho \ll 1$, $\tilde{K}(a') \simeq K(a')$, where $K(a')$ is defined in (3.6) and, in analogy with (3.5),

$$\pi^*(\rho) \simeq \rho[1 + (a - a')\, K(a')]^{-1} \qquad (\rho \ll 1) \tag{3.9}$$

[7] Similar expressions are obtained for the resistivity of a two-dimensional anisotropic network in the effective medium approximation (Ref. 16).

Thus for a very low density ρ of scatterers, the asymptotic occupation probabilities of the scattering sites and thus the diffusion coefficients are identical for periodic[8] and random arrangements of scatterers with identical densities ρ. In view of the remarks in the previous paragraph, the same is true for very high density. The equality of the diffusion coefficients for low density of scatterers for both periodic and random arrangements is in agreement with the analogous Rayleigh–Maxwell result for the effective conductivity for a composite material of conducting disks embedded in a medium of a different conductivity (see Ref. 4).

It is interesting to note that analogous results for periodic and random arrangements of defects were obtained by Argyrakis,[17] who performed Monte Carlo simulations for a two-dimensional lattice, where a fraction ρ of the sites together with the corresponding bonds are deleted. He found a pronounced difference in the expected number S_n of distinct sites visited between a periodic and random arrangement with the same fraction ρ of deleted sites. However, in the limit as ρ goes to zero, the random and periodic results for S_n were found to tend to the same values.

4. DISCUSSION

In this paper we have studied random walks on a number of inhomogeneous lattices and concentrated on the question whether the asymptotic random walk properties (diffusion coefficients in particular) depend explicitly on the spatial arrangement of the inhomogeneities (scatterers, impurities, etc.) or only on their density. Two distinct types of behavior were found.

(i) In the case of the sparsely periodic lattice (Sections 2.1 and 2.2) all the distinct periodic arrangements of columns with the same density give the same answer for the diffusion coefficients. A simple argument based on the Law of large numbers was subsequently used to demonstrate that random arrangements of columns in the unit cell, with the same density as the periodic arrangements, yield in the limit of an infinitely large unit cell diffusion coefficients identical to those of the periodic case. This argument is of a general nature and applies to any system where all the periodic arrangements with identical density of inhomogeneities give rise to identical values for the diffusion coefficients.[9]

[8] Although we only considered the case of a square unit cell, this result also holds for a rectangular unit cell of size $m \times n$ with one scatterer, as long as both m and n are large.

[9] The procedure of going to an infinite unit cell while keeping the density of inhomogeneities constant corresponds, of course, to the standard statistical mechanics procedure of going to the thermodynamic limit. This permits one to prove or demonstrate (depending upon

At this point we cite a few examples from the literature where an analogous situation prevails. The first is the one-dimensional waiting time Lorentz model, already referred to in I, where the diffusion coefficient is identical for lattices with fixed and with random intervals between scatterers for a given density of scatterers.[18,25]

Another example is the continuous time random walk model with isotropic transition probabilities but site-specific waiting time distributions, the Random jump rate model.[19] That is, at every site β the walker jumps with equal probability to one of the neighboring sites but the time between jumps is distributed according to waiting time distributions $\{\psi_\beta(t)\}$ differing from site to site. It can easily be shown from the results in I that for a periodic arrangement of the sites the diffusion coefficient D associated with the total mean square displacement is given by (all lattice constants are assumed to be unity)

$$D = \left(m^{-1} \sum_{\beta=1}^{m} \tau_\beta \right)^{-1} \tag{4.1}$$

where m is the number of sites per unit cell and τ_β is the mean waiting time at site β, i.e., $\tau_\beta = \int_0^\infty dt\, \psi_\beta(t) t$. This result, which is valid in any dimension d, was first derived by Haus *et al.*[20] for the case of exponential waiting time distributions. From (4.1) it is evident that for this periodic case the diffusion coefficient does not depend upon the spatial arrangement of the m mean waiting times $\{\tau_\beta\}$ in the unit cell. The case where the mean waiting time at every site of the infinite lattice is independently chosen from a random distribution can be handled as in Section 2.2.

(ii) The second type of behavior was found in the brick lattice of Section 2.3 and the anisotropic scatterer model of Section 3. Here, different periodic arrangements with identical density of vertical bonds and scatterers, respectively, do not yield the same answer for the diffusion coefficients. Such lattices are prototypes of a wide class of multistate lattices where macroscopic transport properties, such as diffusion coefficients, for random arrangements of inequivalent sites differ from those obtained for periodic arrangements with the same density. Another example in this class is the Random barrier model[19] in dimension $d \geqslant 2$.[10] Here one assigns

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whether the author/reader is a mathematician or a physicist) various properties of infinite systems. The validity of statistical mechanics in the description of physical systems depends on the identity of the results for large, finite systems with those of the corresponding infinite system. Our results, although based on going to the limit of infinite systems, should thus apply to the large, *finite* systems which are the substrates of physics and chemistry.

[10] Only in the one-dimensional case is the diffusion coefficient independent of arrangement.[21-23]

transition rates at random to the *bonds* of a lattice, independent of all the other bonds, such that the rate W_{ij} associated with the bond connecting sites i and j is symmetric, i.e., $W_{ij} = W_{ji}$. The percolation problem is obtained if any of the rates takes the value zero with a positive probability.[15] For some rigorous results in the case of positive but bounded rates, see also Ref. 24.

From our results it is clear that case (i) above is the exception rather than the rule. For the lattices in class (ii) one has to go to very large unit cells, which are macroscopically homogeneous (e.g., realizations of a stationary ergodic or random distribution of inequivalent sites), before the random walk properties *can* become independent of the particular arrangement of the sites. Additional conditions which have to be imposed for this so-called "self-averaging" to happen were discussed by Anshelevich and Vologodskii for one-dimensional lattices.[26] Similar conditions for higher-dimensional lattices do not yet seem to be known. Left open is the question as to a *general* characterization of the types of inequivalent sites (i.e., anisotropic scatterers, partial traps, etc.) which give rise to (i) and (ii), respectively. It would obviously be useful to have an *a priori* knowledge of this so that one would know under what conditions asymptotic random walk properties on randomly inhomogeneous lattices can be calculated via periodic inhomogeneous lattices.

A final remark concerns the usefulness of the approach developed in I and this paper for the calculation of random walk properties such as diffusion coefficients in a random environment. It is clear that in the majority of cases an analytical calculation of these properties is still not feasible. However, based on the results of I and this paper, the following *numerical* procedure suggests itself, which should be more efficient that the usual Monte Carlo simulations. Construct a finite unit cell with randomly chosen sites/bonds and calculate numerically and exactly the diffusion coefficients by the formulas derived in I. This requires only matrix computations. Then add one site/bond to the lattice, chosen at random from the probability distribution of sites/bonds. Again compute the diffusion coefficients and repeat this procedure until convergence is observed. A similar procedure has recently been developed to calculate the resistance of a random network.[27]

Note Added in Proof. Some of the results of Part I were obtained independently by A. Krámli and D. Szász, *Z. Wahrscheinlichkeitstheorie verw. Gebiete* **52**:309–319 (1980). We thank the referee for bringing this reference to our attention.

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APPENDIX A

In this appendix, we derive Eqs. (2.19) and (2.20). The unit cell of the deformed lattice is indicated in Fig. 4, which contains three lattice constants a, b, c and two angles α and β, which are interrelated. First of all, from the geometry one deduces that

$$\frac{\sin\alpha}{\sin\beta} = K\frac{b}{a} \tag{A1}$$

where

$$K = \frac{m-1}{k-(m-1)} \tag{A2}$$

Secondly, from the requirement that the walk be locally unbiased in all directions, we obtain

$$a\cos\alpha = b\cos\beta \tag{A3}$$

$$a\sin\alpha + b\sin\beta = c \tag{A4}$$

The occupation probabilities of the sites are the same as for the original lattice, Eq. (2.14).[9] The single-step variances are

$$\langle x^2\rangle_\beta = \tfrac{2}{3}a^2\cos^2\alpha, \qquad \langle y^2\rangle_\beta = \tfrac{2}{3}b^2\sin^2\beta(1+K+K^2), \qquad \beta = 1,\ m,\ k+1,\ k+m \tag{A5a}$$

$$\langle x^2\rangle_\beta = a^2\cos^2\alpha, \qquad \langle y^2\rangle_\beta = K^2b^2\sin^2\beta, \qquad \beta = m+1,\ldots,\ k;\quad k+m+1,\ldots,\ 2k \tag{A5b}$$

$$\langle x^2\rangle_\beta = a^2\cos^2\alpha, \qquad \langle y^2\rangle_\beta = b^2\sin^2\beta, \qquad \beta = 2,\ldots,\ m-1;\quad k+2,\ldots,\ k+m-1 \tag{A5c}$$

where we have used (A1)–(A4). By using (2.14) and (A5), we find

$$2D'_x = \sum_\beta \langle y^2\rangle_\beta \pi_\beta = a^2\cos^2\alpha\,\frac{k}{k+1} \tag{A6}$$

and

$$D'_y = \frac{1}{k+1} b^2 \sin^2 \beta [2(1+K+K^2) + (k-m) K^2 + m - 2] \tag{A7}$$

The off-diagonal elements of D′ are zero. From the geometry of the original lattice we find,

$$\mathsf{A} = \begin{pmatrix} k & 0 \\ 0 & 2 \end{pmatrix} \tag{A8}$$

and similarly for the deformed lattice,

$$\mathsf{A}' = \begin{pmatrix} ka \cos \alpha & 0 \\ 0 & 2b \sin \beta(m+K) \end{pmatrix} \tag{A9}$$

From (2.18) we then obtain by using (A6)–(A9),

$$2D_x = \frac{k}{k+1} \tag{A10}$$

and

$$2D_y = \frac{1}{k+1} \frac{1}{\gamma} \tag{A11}$$

where

$$\gamma = \frac{(m+K)^2}{2K + K^2(k-m+2) + m} = \frac{(1+K)^2 + Kk}{(1+K)^2} \tag{A12}$$

Inserting (A2) in (A12), we arrive at the desired results (2.19) and (2.20).

APPENDIX B

Here we derive the low-density results (3.5)–(3.8) for two-dimensional lattices with anisotropic scatterers. We start with the random case. The square unit cell consists of $m \times m$ sites, labeled by (ij), where i indicates the row on which the site is located and j the column ($i, j = 0, 1, \ldots, m-1$). The elements $\langle ij| T |i'j' \rangle$ of the transition matrix T describing the embedded Markov Chain on the m^2 internal states, are given by[11]

$$\begin{aligned} \langle ij| T |i'j' \rangle = {} & \varepsilon_{i'j'} \{\delta_{i',i+1} + \delta_{i',i-1}\} \delta_{j,j'} \\ & + (\tfrac{1}{2} - \varepsilon_{i'j'}) \delta_{i,i'} \{\delta_{j',j+1} + \delta_{j',j-1}\} \end{aligned} \tag{B1}$$

[11] The symbol $\delta_{i,j}$ denotes the Kronecker δ function.

where

$$\varepsilon_{i'j'} = a' + (a - a')\,\alpha_{i'j'} \tag{B2}$$

and

$$\alpha_{i'j'} = \begin{cases} 1 & \text{with probability } v \\ 0 & \text{with probability } 1 - v \end{cases} \tag{B3}$$

That is, with probability v the transition probabilities are a and $\frac{1}{2} - a$ in the horizontal and vertical direction, respectively, ("scatterers"), and with probability $1 - v$ the transition probabilities are a' and $\frac{1}{2} - a'$ ("regular sites"). The quantity to be determined is the occupation probability $\overline{\pi^*}(v)$ of the infinite random lattice. If we denote the components of the right eigenvector $\boldsymbol{\pi}$ of (B1) belonging to the eigenvalue $\lambda_0 = 1$ by π_{ij} $(ij = 0, 1, \ldots, m-1)$, then the *average* occupation probability of the scatterers for finite m is given by

$$\overline{\pi^*}(m, v) = \sum_{i,j} \overline{\alpha_{ij} \pi_{ij}} \tag{B4}$$

where the bars denote averages over the random variables α_{ij}. The desired quantity $\overline{\pi^*}(v)$ is then finally obtained by taking the limit of an infinite unit cell,[12]

$$\overline{\pi^*}(v) = \lim_{m \to \infty} \overline{\pi^*}(m, v) \tag{B5}$$

The eigenvector $\boldsymbol{\pi}$ of (B1) will now be determined by perturbation theory in powers of v, i.e., we assume that $v \ll 1$. The unperturbed matrix T_0 is defined by its matrix elements,

$$\begin{aligned} \langle ij|\, T_0\, |i'j'\rangle = {} & a' \{\delta_{i',i+1} + \delta_{i',i-1}\}\, \delta_{j,j'} \\ & + \{\tfrac{1}{2} - a')\, \delta_{i,i'}\, \{\delta_{j',j+1} + \delta_{j',j-1}\} \end{aligned} \tag{B6}$$

or, by introducing Kronecker products, indicated by the symbol $\otimes$,

$$\mathsf{T}_0 = a' \mathsf{R} \otimes \mathsf{1} + (\tfrac{1}{2} - a') \mathsf{1} \otimes \mathsf{R} \tag{B7}$$

where the m-dimensional square matrix R is given by

$$\mathsf{R} = \begin{pmatrix} & 1 & & & 1 \\ 1 & & 1 & & \\ & 1 & & \ddots & \\ & & 1 & & \ddots \\ & & & \ddots & 1 \\ 1 & & & 1 & \end{pmatrix} \tag{B8}$$

[12] For the validity of this procedure, see Section 2.2.

and 1 is the m-dimensional unit matrix. The perturbation T_1 is defined by

$$\langle ij|\ T_1|i'j'\rangle = \alpha_{i'j'}(a-a')[\{\delta_{i',i+1}+\delta_{i',i-1}\}\ \delta_{j,j'} - \delta_{i,i'}\{\delta_{j',j+1}+\delta_{j',j-1}\}] \tag{B9}$$

The eigenvalue equation to be solved is

$$(\mathsf{T}_0+\varepsilon\mathsf{T}_1)\boldsymbol{\pi}=\boldsymbol{\pi} \tag{B10}$$

where we have inserted the parameter ε to keep track of the orders of T_1. It is convenient to diagonalize T_0 first, by introducing the Fourier matrix F, with components

$$F_{i'j'}=m^{-1/2}z^{i'j'} \qquad (i', j'=0, 1,..., m-1) \tag{B11}$$

where

$$z=\exp\left(\frac{2\pi i}{m}\right) \tag{B12}$$

and the property that $\mathsf{F}^{-1}=\mathsf{F}^*$. Equation (B10) is now transformed to the equivalent equation

$$(\mathsf{M}_0+\varepsilon\mathsf{M}_1)\mathbf{x}=\mathbf{x} \tag{B13}$$

where

$$\mathbf{x}=(\mathsf{F}^*\otimes\mathsf{F}^*)\boldsymbol{\pi} \tag{B14}$$

$$\mathsf{M}_0=(\mathsf{F}^*\otimes\mathsf{F}^*)\,\mathsf{T}_0(\mathsf{F}\otimes\mathsf{F})=a'\boldsymbol{\Lambda}\otimes\mathsf{1}+(\tfrac{1}{2}-a')\mathsf{1}\otimes\boldsymbol{\Lambda} \tag{B15}$$

and

$$\mathsf{M}_1=(\mathsf{F}^*\otimes\mathsf{F}^*)\,\mathsf{T}_1(\mathsf{F}\otimes\mathsf{F}) \tag{B16}$$

The matrix $\boldsymbol{\Lambda}=\mathsf{F}^*\mathsf{R}\mathsf{F}$ in (B15) is diagonal with elements

$$\lambda_j=2\cos\left(\frac{2\pi j}{m}\right) \qquad (j=0, 1,..., m-1) \tag{B17}$$

The matrix elements of M_0 and M_1 are

$$\langle ij|\ M_0\ |i'j'\rangle=\delta_{ii'}\delta_{jj'}[a'\lambda_i+(\tfrac{1}{2}-a')\,\lambda_j] \tag{B18}$$

and

$$\langle ij|\ M_1\ |i'j'\rangle=\frac{1}{m^2}(a-a')\{\lambda_i-\lambda_j\}\,\hat{\alpha}_{i-i',j-j'} \tag{B19}$$

where

$$\hat{\alpha}_{i,j} = \sum_{k,l} (z^*)^{ik} (z^*)^{jl} \alpha_{kl} \tag{B20}$$

The solution of (B13) is written as a power series in ε,

$$\mathbf{x} = \mathbf{x}_0 + \varepsilon \mathbf{x}_1 + \varepsilon^2 \mathbf{x}_2 + \cdots$$

The unperturbed solution, properly normalized, is easily found to be

$$(\mathbf{x}_0)_{ij} = \frac{1}{m} \delta_{i,0} \delta_{j,0} \tag{B21}$$

The complete solution $\mathbf{x}$ can now be written

$$\mathbf{x} = \frac{1}{m} \begin{pmatrix} 1 \\ \tilde{x} \end{pmatrix} \tag{B22}$$

where the $(m^2 - 1)$-dimensional vector $\tilde{x}$ is of $O(\varepsilon)$:

$$\tilde{x} = \varepsilon \tilde{x}_1 + \varepsilon^2 \tilde{x}_2 + \cdots \tag{B23}$$

The coefficient of ε^n obeys the equation [$\tilde{1}$ is the unit matrix of dimension $(m^2 - 1)$]

$$(\tilde{M}_0 - \tilde{1})\, \tilde{x}_n + \tilde{M}_1 \tilde{x}_{n-1} = 0 \qquad (n \geqslant 2) \tag{B24}$$

where the matrices $\tilde{M}_0$ and $\tilde{M}_1$ are obtained from M_0 and M_1 by deleting the first row and first column. The solution of (B24) is

$$\tilde{x}_n = [-(\tilde{M}_0 - \tilde{1})^{-1} \tilde{M}_1]^{n-1} \tilde{x}_1 \qquad (n \geqslant 2) \tag{B25}$$

and thus

$$\tilde{x} = \sum_{n=1}^{\infty} \varepsilon^n [-(\tilde{M}_0 - \tilde{1})^{-1} \tilde{M}_1]^{n-1} \tilde{x}_1 \tag{B26}$$

The vector $\mathbf{x}_1$ obeys

$$(\mathsf{M}_0 - 1)\, \mathbf{x}_1 + \mathsf{M}_1 \mathbf{x}_0 = 0 \tag{B27}$$

from which, by using (B18) and (B21), one obtains

$$(\tilde{x}_1)_{ij} = -\left[a' \lambda_i + \left(\frac{1}{2} - a' \right) \lambda_j - 1 \right]^{-1} \frac{1}{m^2} (a - a')(\lambda_i - \lambda_j)\, \hat{\alpha}_{ij} \tag{B28}$$

Using (B14), (B11), and (B22), we can write the average occupation probability (B4) as

$$\overline{\pi^*} = \frac{1}{m}\sum_{ij}\sum_{i'j'}\overline{\alpha_{ij} z^{ii'} z^{jj'} \frac{1}{m}\left[\delta_{i',0}\delta_{j',0} + (1-\delta_{i',0}\delta_{j',0})(\tilde{x})_{i'j'}\right]}$$

$$= v + \frac{1}{m^2}\sum_{ij}{\sum_{i'j'}}' \overline{\alpha_{ij} z^{ii'} z^{jj'} (\tilde{x})_{i'j'}} \tag{B29}$$

where we have used that $\overline{\alpha_{ij}} = v$, and where the prime on the summation sign indicates that the term with $i' = 0$, $j' = 0$ has to be omitted. From (B18)–(B20), (B26), and (B28), we have (putting $\varepsilon = 1$),

$$\overline{\pi^*} = v + \sum_{n=1}^{\infty}\left[(-)^n \frac{1}{m^2}\left(\sum_{ij}\sum_{k_1 l_1}\cdots\sum_{k_n l_n}\right)\left({\sum_{k_1' l_1'}}'\cdots{\sum_{k_n' l_n'}}'\right)\gamma_{k_1' l_1'}\cdots\gamma_{k_n' l_n'}\right.$$
$$\times z^{ik_1'} z^{jl_1'} z^{-k_1(k_1'-k_2')} z^{-l_1(l_1'-l_2')}\cdots z^{-k_{n-1}(k_{n-1}'-k_n')} z^{-l_{n-1}(l_{n-1}'-l_n')}$$
$$\left.\times z^{-k_n k_n'} z^{-l_n l_n'} \overline{\alpha_{ij}\alpha_{k_1 l_1}\cdots\alpha_{k_n l_n}}\right] \tag{B30}$$

where

$$\gamma_{ij} = \left[a'\lambda_i + \left(\frac{1}{2} - a'\right)\lambda_j - 1\right]^{-1} \frac{(a-a')}{m^2}(\lambda_i - \lambda_j) \tag{B31}$$

Since we cannot evaluate (B30) exactly, we confine ourselves here to all terms in the right-hand side of (B30) which are linear in v. These are the terms where $i = k_1 = \cdots = k_n$, $j = l_1 = \cdots = l_n$, for $\overline{\alpha_{ij}^n} = \overline{\alpha_{ij}} = v$. All other terms are of order v^2.

Summing all these terms linear in v, we obtain

$$\overline{\pi^*} = v + v\sum_{n=1}^{\infty}(-)^n \frac{1}{m^2}\sum_{ij}{\sum_{k_1' l_1'}}'\cdots{\sum_{k_n' l_n'}}' \gamma_{k_1' l_1'}\cdots\gamma_{k_n' l_n'} + O(v^2)$$

$$= v\left[1 + {\sum_{k,l}}' \gamma_{kl}\right]^{-1} + O(v^2) \tag{B32}$$

If we finally take the limit $m \to \infty$, we get

$${\sum_{k,l}}' \gamma_{kl} \to (a-a')\,K(a') \tag{B33}$$

where $K(a')$ is given by (3.6) and (B32) reduces to the result (3.5) as had to be shown.

In the case of a nonrandom periodic lattice with one scatterer at the origin (0, 0) of the unit cell, we can repeat the same derivation as above, but now with

$$\alpha_{ij} = \begin{cases} 1 & \text{if } (ij) = (0,0) \\ 0 & \text{otherwise} \end{cases} \tag{B34}$$

and one arrives at the exact result

$$\pi^* = \frac{1}{m^2}\left[1 + \sum_{k,l}{}' \gamma_{kl}\right]^{-1} \tag{B35}$$

which is (3.7).

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