Density expansion of transport properties on 2D site-disordered lattices: I. General theory

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Abstract. A systematic kinetic theory is developed for calculating transport properties on 2D lattices with random site impurities in concentration c, which can be modelled by hopping models. Our main results are expressions in terms of lattice sums for the static and frequency-dependent conductivity and for the velocity autocorrelation function.

1. Introduction

In this paper we develop a new method based on kinetic theory (Dorfman 1975) for obtaining systematic density expansions of transport properties in disordered lattices and percolation problems, and obtain new results for static and frequency-dependent transport properties, *exact to quadratic order in the concentration c of impurities.*

To describe the trajectory of a particle moving in a static environment of scatterers we have to specify the dynamics at the microscopic level, which contains the full complications of the Liouville equation. The art in kinetic theory is to construct a model that is mathematically tractable and physically realistic enough to capture some of the essential mechanims. Thus, one simplifies from fluids with general pair interactions to Lorentz models where one particle moves with a constant speed in a random array of fixed hard scatterers. More drastically, one replaces the ballistic trajectories between successive collisions by a random walk, and puts the random walker on a lattice, and one may restrict his jumps to nearest-neighbour (NN) hops. The model considered here is one of the most simplified choices according to these options.

However, the resulting models still have an enormous richness, which depends on the structure and connectedness of the underlying disordered lattice. They are known, e.g., as the theory of random resistor networks, conduction–percolation theory, ants in a labyrinth, etc.

In these kinetic models we have lost the feature that the mean free path λ_0 at low density of scatterers is inversely proportional to their density. In lattice hopping the 'mean free path' is simply the lattice distance. The drastic consequences of this simplification can be seen by comparing the hopping models with fluids and Lorentz gases. The repeated ring collisions in, say, a 2D or 3D Lorentz gas (Dorfman 1975) give a contribution to the transport coefficients growing as $\ln \lambda_0 \sim \ln(1/c)$ and yields a relative correction, $c \ln c$ or $c^2 \ln c$, respectively, to the low density Boltzmann value

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of the diffusion coefficient. Therefore a *virial expansion* of the diffusion coefficient in powers of the density does not exist.

The occurrence of these logarithmic terms has blocked the further development of a *systematic kinetic theory* for calculating transport properties in moderately dense systems. One has developed ring kinetic equations (see the review by Dorfman and van Beijeren 1977) to account for the memory effects induced by sequences of correlated binary collisions (recollisions, repeated ring collisions), which explain the $c \ln c$ terms and the long time tails of the time correlation functions at low densities, but do not give quantitative predictions for transport coefficients in moderately dense systems.

In our hopping models the uncorrelated binary collisions described by the Boltzmann equation are replaced by an ordinary random walk (RW), and the sequences of correlated binary collisions correspond to sequences of repeated visits of the RW to single impurities, pairs of impurities, etc (Ernst 1986). For such systems the mean free path is simply the lattice distance and a systematic density expansion of transport coefficients can be developed. Furthermore, long time tails are still present (Frenkel 1987) and Nieuwenhuizen *et al* (1986, hereafter referred to as I).

Density expansions have been made of static properties of random lattices, such as the spectral properties of mass-disordered harmonic lattices (Lifshitz 1964). In percolation problems p expansions ($p = 1 - c$) have been obtained for percolation probabilities (Gaunt and Sykes 1983) and connectedness correlation functions and their moments, through a mapping on the q-state Potts model with $q = 1$ (Fish and Harris 1978). However, systematic density expansions of non-equilibrium properties beyond the single impurity approximation (Izyumov 1966, Harris and Kirkpatrick 1977, Roerdink and Shuler 1985, Roerdink 1985) are virtually non-existent.

There exists an enormous literature on diffusion and conduction in random systems, Lorentz gases (Hauge 1974), hopping models or random walks (RW) on lattices with random disorder in bonds or sites (for a review see Haus and Kehr 1987) and termite problems (Hong *et al* 1986), dynamic percolation, random resistor networks (Kirkpatrick 1973), etc. Most theoretical studies and computer simulations have concentrated on percolation problems in the close vicinity of the percolation threshold. The standard methods to calculate transport properties for general impurity concentration are effective medium type approximations (Kirkpatrick 1973, Watson and Leath 1974, Odagaki *et al* 1983, Odagaki 1986, Garbozci and Thorpe 1986), transfer matrix methods (Herrmann *et al* 1984), Monte Carlo simulations (Argyrakis and Kopelman 1980, Pandey and Stauffer 1983) and real space renormalisation group calculations (Stinchcombe and Watson 1976, Wilkinson *et al* 1983, Luck 1985, Hong *et al* 1986, Costa *et al* 1986).

Lifshitz and Stepanova (1956) have developed a systematic method for expanding properties of disordered lattices in powers of the concentration c of impurities. We follow partly their method which yields for the coefficient of c^l exact expressions in the form of 'cluster integrals' (lattice sums) involving l impurities. However, the Lifshitz 'trace method' for evaluating these cluster integrals is applicable to the density of states and further *equilibrium properties*, such as the free energy, that can be expressed as integrals over the density of states (Lifshitz 1964, § 6.1). The cluster integrals for *non-equilibrium properties*, such as moments of displacement, velocity autocorrelation function (VACF), etc, are more complicated objects, for which no manageable extension of the Lifshitz trace method seems feasible.

However, we will show here that these non-equilibrium cluster integrals can be exactly evaluated using kinetic theory methods. For bond disordered lattices such

calculations are rather simple (Ernst *et al* 1987). For site disorder, to be considered in this paper, such calculations are unfortunately rather complicated.

The reason for presenting our calculations is that this model has been studied frequently in the literature (diffusion coefficient $D(c)$, VACF) using computer simulations and approximate theories. Because of many conflicting results in the literature, as extensively discussed in I, there is clearly a great demand for exact results. For the percolation case ($\sigma = 0$) exact results up to $O(c^2)$ for the diffusion coefficient $D(c) = D_0(1 + \alpha_1 c + \alpha_2 c^2)$ and for the long time tail of the VACF $\varphi(t) = \langle v_x(t)v_x(0)\rangle$, namely $t^2\varphi(t) \sim \eta_0 c + \eta_1 c^2 (t \to \infty)$, have been reported in I. Results for $D(c)$ and VACF up to $O(c^2)$ for general σ values will be obtained in this and the next paper in the series.

The plan of this paper is as follows. In § 2 we introduce the hopping model on a square lattice with random site impurities and show the close connection between the continuous (CTRW) and the discrete (DTRW) time random walk. In § 3 we develop a density expansion and introduce a T matrix resummation to evaluate the required cluster integrals. To simplify our results we shall heavily use the symmetry properties of Green functions on the square lattices which are summarised in appendix 2. From the response function we extract in § 4 expressions for static and frequency-dependent transport coefficients, exact to $O(c^2)$, which are the main results of this paper. These expressions were the starting point of the percolation results reported in I.

2. Continuous and discrete time random walk

The system to be studied is a square lattice with unit lattice distance, having N sites, labelled $n = \{n_x, n_y\}$ and obeying periodic boundary conditions. A fraction c of the sites in the host lattice, chosen at random, is replaced by impurities or scatterers and we study random walks on this random lattice.

In the continuous time random walk (CTRW) the random walker (RW) pauses at every site n during a random waiting time τ which is exponentially distributed as $\exp(-\tau/\tau_0)$ with τ_0 the average waiting time. Units are chosen such that $\tau_0 = 1$. The master equation describes the time evolution of the probability distribution $p_n(t)$ as

$$\dot{p}_n = \tfrac{1}{4}\sum_\rho (\psi_n p_{n+\rho} - \psi_{n+\rho} p_n) \equiv -(\hat{L}p)_n \tag{2.1}$$

where ρ denotes one of the four NN lattice vectors: $\rho = \{\pm 1, 0\}$ or $\{0, \pm 1\}$. We have assigned to every site n a random variable ψ_n, representing the transition probability per unit time for NN hops. It is defined as $\psi_n = 1 - bc_n$, with $b \equiv 1 - \sigma \leq 1$, where $c_n = 1$ for an impurity site and $c_n = 0$ for an impurity free site, namely

$$c_n = \begin{cases} 1 \\ 0 \end{cases} \quad \text{with probability} \quad \begin{matrix} c \\ 1 - c. \end{matrix} \tag{2.2}$$

Here $b < 0$ corresponds to $\sigma > 1$ and $0 < b \leq 1$ corresponds to $1 > \sigma \geq 0$. The master equation admits the (normalised) stationary solution

$$p_n^0 = C\psi_n = \psi_n/(N\langle\psi\rangle) \tag{2.3}$$

with $\langle\psi\rangle = 1 - bc$, where the average $\langle \ldots \rangle$ over the quenched disorder is taken over the probability distribution (2.2) of the set of random variables $\{c_n\}$.

We make some comments on the model. The transition *probability per unit time for NN hops* ψ_n depends in the present site model *only on the site of arrival*.

The jump rate into an impurity site, $\psi_n = 1 - b = \sigma$, may be smaller or larger than the rate into an impurity free site with $\psi_n = \sigma_0 = 1$. For $\sigma = 0$ or $b = 1$ one has the standard site percolation problem with inaccessible impurity sites. At small impurity concentrations one has the model of blind (unbiased) ants hopping on the percolation cluster far above threshold. At low impurity concentration our site problem may also be considered as a lattice Lorentz gas (Hauge 1974), where the ballistic motion of the moving particle has been replaced by the stochastic motion of the RW. For $\sigma \gg 1$ one has 'superconducting' impurities, often referred to as 'termite' models.

One may also study the relaxational modes of (2.1) by letting $p_n(t) = p_n \exp(-Et)$, where E are the eigenvalues (rate constants). This eigenvalue problem is also identical to that for spin wave amplitudes in which NN exchange integrals are unity and the spin magnetisation equals $\psi_n = 1$ on a host lattice site and $\psi_n = \sigma$ on an impurity site. In this context the model has been used by Harris and Kirkpatrick (1977) to calculate spin wave stiffness constants to $O(c)$ in the impurity concentration.

The transport properties of interest are the diffusion coefficient $D(c)$, the velocity autocorrelation function (VACF) $\varphi(t) = \langle v_x(0)v_x(t)\rangle$ and the frequency-dependent conductivity. They are all related to the mean square displacement $\langle n_x^2\rangle(t)$. One may also consider the Burnett transport coefficients, which are related to the fourth cumulant of the displacements.

The most important moment is the mean square displacement, in terms of which we introduce a time-dependent diffusion coefficient

$$\tilde{D}(t) = \tfrac{1}{2}(\partial/\partial t)\langle n_x^2\rangle(t) = \int_0^t d\tau\,\varphi(\tau). \tag{2.4}$$

Its long time behaviour represents the static diffusion coefficient $\tilde{D}(\infty) = D$. The second derivative of the mean square displacement is the lattice analogue of the VACF.

The macroscopic conductivity is related to the diffusion coefficient through the Einstein relation $\Sigma = \text{constant} \times n_0 D$, where n_0 is the density of carriers in the stationary state proportional to the effective number of accessible sites or (effective) *free volume* $\langle \psi \rangle = 1 - bc$. In our general models the effective free volume of impurity sites may be lower ($\sigma < 1$) or higher ($\sigma > 1$) than that of impurity free sites. In the context of transport in porous media $\langle \psi \rangle$ is referred to as porosity (Guyer 1987).

The relation between conductivity and diffusion coefficient on a lattice with a fraction c of impurity sites is, in appropriate units,

$$\Sigma(c) = \langle \psi \rangle D(c).$$

We further consider a 'frequency-dependent' diffusion coefficient $\Phi(z)$, which is the Laplace transform of the VACF $\varphi(t)$. Its small-z behaviour gives the static diffusion coefficient $D(c) = \Phi(z = 0)$. In a similar fashion we introduce a 'frequency-dependent' conductivity $\Xi(z) = \langle \psi \rangle \Phi(z)$, where the actual AC conductivity of the system is determined by $\Sigma(c, \omega) = \text{Re }\Xi(i\omega)$. A further property of interest is the staying probability or return probability $P_0(t)$ of the RW to its point of origin. All transport properties described above can be expressed in terms of the macroscopic probability for a displacement from m to n in a time t, $P_{n-m}(t)$, *averaged over the quenched disorder*, i.e. averaged over the the probability distribution of the random variables $\{c_n\}$

$$P_{n-m}(t) = \langle p(nt\,|\,m0)p_m^0\rangle \tag{2.5}$$

where the initial positions of the RW are weighted with the stationary solution p_m^0 of the master equation, given in (2.3). The quantity $p(nt\,|\,m0)$ is the solution of the master equation with initial condition $p(n0\,|\,m0) = \delta_{nm}$.

It is of interest to indicate the connection with the discrete time random walk (DTRW) which is particularly relevant because computer simulations are done with discrete time steps.

We consider again a lattice with a random mixture of jump rates $\sigma_0 = 1$ and $\sigma = 1 - b$. In the case $\sigma < \sigma_0 = 1$ (impurity bonds with lower conductivity than that of the host lattice bonds) we set our clock to tick every $1/\sigma_0$ seconds and *normalise the total jumping probability to unity for a site without impurity NN sites*. Let τ denote the number of ticks or of time units. The probability to find the RW at site n after $(\tau + 1)$ time units is then given by

$$p_{n,\tau+1} - p_{n,\tau} = \tfrac{1}{4} \sum_{\rho} [\psi_n p_{n+\rho,\tau} - \psi_{n+\rho} p_{n,\tau}] \equiv -(\hat{L} p_\tau)_n \tag{2.6}$$

which is very similar to (2.1) for the CTRW.

On a uniform lattice there is a simple relationship between the probability distribution and related quantities of the CTRW at time t and those of the DTRW after τ hops of the RW (Haus *et al* 1983). The same relationship holds on a disordered lattice, if the waiting time distribution is the same at each site (Machta 1981)

$$P_n(t) = \sum_{s=0}^{\infty} p_{n,s} \Psi_s(t) = e^{-t} \sum_{s=0}^{\infty} p_{n,s} t^s / s! \tag{2.7}$$

where $\Psi_s(t)$ is the probability for *exactly* s ticks of our clock during a time interval t. The second equality holds, since the waiting time in our model is distributed according to $\exp(-t)$, from where $\Psi_s(t) = (t^s/s!) \exp(-t)$ follows. For the VACF φ_s and the time-dependent diffusion coefficient $\tilde{D}_s$ in the DTRW we find similarly

$$\varphi(t) = \tfrac{1}{2} \varphi_0 \delta_+(t) + e^{-t} \sum_{s=0}^{\infty} \varphi_{s+1} t^s / s!$$

$$\tilde{D}(t) = e^{-t} \sum_{s=0}^{\infty} \tilde{D}_{s+1/2} t^s / s! \tag{2.8}$$

where $\delta_+(t)$ is a Dirac delta function, normalised to unity on the time interval $[0, \infty)$. These relations have been used by Frenkel (1987) to transform his results for the discrete time VACF φ_τ, and diffusion coefficient D_s obtained from computer simulation, into the VACF φ_t and diffusion coefficient $\tilde{D}(t)$ with a continuous time t. The latter quantity can be compared directly with our theoretical predictions. From here on we restrict ourselves to the continuous time version.

For our purpose it is more convenient to write the master equation for the CTRW (2.1) in the form

$$\dot{p}_n = -\tfrac{1}{4} \sum_{\rho} (E_\rho^{-1} - 1) \psi_n E_\rho p_n$$

$$\equiv -\sum_{m} \hat{L}_{nm} p_m \equiv -(Lp)_n \tag{2.9}$$

where $E_\rho f(n) = f(n + \rho)$. We are interested in the moments of displacement $\langle (n_x - m_x)^l \rangle = \Sigma_{n,m} (n_x - m_x)^l \langle p(nt; m0) \rangle$ where $\langle p(nt; m0) \rangle$ is the time-dependent probability distribution $p(nt; m0) = p(nt|mo) p_m(0)$ (see (2.5)), and the initial distribution $p_m(0)$ is always taken to be the stationary solution (2.3), namely $p_m(0) = p_m^0$. Thus we have for the macroscopic distribution of displacements

$$P_{n-m}(t) = \langle p(nt; m0) \rangle = \langle (e^{-tL})_{nm} \psi_m \rangle / N \langle \psi \rangle. \tag{2.10}$$

The response function is the generating function for the (Laplace transformed) moments of displacement and follows from (2.10) as

$$F(q, z) = \sum_{n,m} \exp[iq(n-m)]\langle(z+\hat{L})^{-1}_{nm}\psi_m\rangle/N\langle\psi\rangle$$
$$= \langle(z+L)^{-1}\Psi\rangle_{qq}/\langle\psi\rangle \tag{2.11}$$

where $\Psi_{qq'}$ is the Fourier representation of the diagonal matrix $\hat{\Psi}_{nm} = \psi_n\delta_{nm}$. In general the matrix $A_{qq'}$ is defined in terms of its coordinate representation $\hat{A}_{nm}$ as

$$A_{qq'} = N^{-1} \sum_{n,m} \exp(inq)\hat{A}_{nm}\exp(-imq') \tag{2.12}$$

where q is a reciprocal lattice vector restricted to the first Brillouin zone (IBZ). We frequently use the orthogonality relations

$$N^{-1}\sum_n \exp[in(q-q')] = \delta_{qq'} \qquad N^{-1}\sum_q \exp[iq(n-m)] = \delta_{nm}. \tag{2.13}$$

Here and in the following all q sums extend over the IBZ. Derivatives of the response function yield the mean square displacement and higher moments, for example

$$\langle n_x^2\rangle(z) = -(\partial/\partial q_x)^2 F(q, z)|_{q=0}. \tag{2.14}$$

3. Density expansions

To derive a density expansion we follow Lifshitz and Stepanova's method and expand the response function in (2.11) in powers of the concentration c of impurities:

$$\langle\psi\rangle F(q, z) = \langle H(q, z)\rangle = f_0(q, z) + cf_1(q, z) + c^2f_2(q, z) + \dots \tag{3.1}$$

where $\langle H\rangle$ is actually the average of

$$H(q, z) \equiv ((z+L)^{-1}\Psi)_{qq} \tag{3.2}$$

over the quenched disorder. Then

$$f_0(q, z) = H_0$$
$$f_1(q, z) = [\partial\langle H\rangle/\partial c]_{c=0} = N(\langle H_1\rangle - H_0)$$
$$f_2(q, z) = \tfrac{1}{2}[\partial^2\langle H\rangle/\partial c^2]_{c=0} = \tfrac{1}{2}N^2(\langle H_2\rangle - 2\langle H_1\rangle + H_0) \tag{3.3}$$

where $f_l(q, z)$ is simply proportional to the lth finite difference of $\langle H_m\rangle$. Let H_m be the (unaveraged) response function of a lattice containing precisely m impurities at the fixed sites $\{n_1, n_2, \dots n_m\}$ and let

$$h_m(n_1, n_2 \dots n_m) = N[H_m(n_1n_2\dots n_m) - H_0] \tag{3.4}$$

be the addition to the unperturbed response function H_0.

For an asymptotically large number of sites N, $h_1(n_1) = h_1$ is independent of n_1 and $h_2(n_1, n_2) = h_2(n_2 - n_1)$, while $h_2(n) \to 2h_1$ as $n \to \infty$. Next, we write (3.3) in terms of h_m and write out the averages in (3.3), observing that the probabilities for finding different impurities at different sites are uncorrelated in the present model. This immediately yields for f_l the following cluster integrals (lattice sums):

$$f_0 = H_0 \qquad f_1 = h_1$$
$$f_2 = \tfrac{1}{2}\sum(h_2(n) - 2h_1) \tag{3.5}$$
$$f_3 = \frac{1}{3!}\sum'(h_3(n, m) - h_2(n) - h_2(m) - h_2(n - m) + 3h_1).$$

The cluster integrals in (3.5) have the same structure as the *l*-tuple collision integrals in kinetic theory. For deterministic models (fluids, Lorentz gases) higher cluster integrals start to diverge as $z \to 0$, causing a breakdown of the density expansion of transport coefficients and leading to the appearance of $\rho^l \ln \rho$ terms in their small density (ρ) behaviour (Dorfman and van Beijeren 1977). In our stochastic models of RW on disordered lattices the cluster integrals converge as $z \to 0$, but in the presence of random traps small-z divergences start to reappear again (Felderhof *et al* 1982).

In order to evaluate these quantities we follow the standard methods of kinetic theory and introduce a binary collision expansion of the m impurity operator $H_m = ((z + L_m)^{-1} \Psi_m)_{qq}$ where L_m and Ψ_m are as L and Ψ with $c_n = 1$ for $n \in \{n_1, n_2, \ldots, n_m\}$ and $c_n = 0$ otherwise. For convenience of notation we drop the subscript m, unless explicit specification of the impurity sites is required.

To proceed we split L and Ψ in (3.2) into a random part, linear in c_n, and into a sure part, independent of c_n, and referring to the uniform host lattice $L = L^0 - \delta L$ and $\Psi = 1 - \delta\Psi$, where 1 is a unit matrix and

$$L^0_{qq'} = \omega(q)\delta_{qq'} \qquad \delta\Psi_{qq'} = bC_{qq'}$$
$$\delta L_{qq'} = b[\omega(q') - \omega(q - q')]C_{qq'}. \tag{3.6}$$

In the equations above, $C_{qq'}$ is the Fourier representation of the diagonal matrix $\hat{C}_{nm} = c_n\delta_{nm}$ and

$$\omega(q) = \tfrac{1}{4}\sum_\rho [1 - \exp(iq\rho)] = 1 - \tfrac{1}{2}c_1 - \tfrac{1}{2}c_2. \tag{3.7}$$

Here and in the following we use the shorthand notation

$$c_1 = \cos q_x \qquad c_2 = \cos q_y \qquad s_1 = \sin q_x \qquad s_2 = \sin q_y. \tag{3.8}$$

A formal perturbation expansion of $H(q, z)$ in (3.2) can be obtained by using $(z + L^0 - \delta L)^{-1} = g \sum_{l=0}^\infty (\delta L g)^l$ where

$$g_{qq'} = g(q)\delta_{qq'} = (z + \omega(q))^{-1}\delta_{qq'}. \tag{3.9}$$

Insertion of this expansion in (3.4) in combination with $H_0 = (z + L_0)^{-1}_{qq} = g(q)$ yields after some rearrangements

$$H(q, z) = g(q) + \left(g \sum_{l=0}^\infty (\delta L g)^l \Lambda g\right)_{qq} \tag{3.10}$$

where

$$\Lambda_{qq'} = \delta L_{qq'} - \delta\Psi_{qq'}(z + \omega(q')) = -b(z + \omega(q - q'))C_{qq'}. \tag{3.11}$$

In view of the developments in later sections we want to factorise the q and q' dependence of $\delta L_{qq'}/C_{qq'}$ and $\Lambda_{qq'}/C_{qq'}$. For the square lattice this can be done conveniently using the following 5×5 matrix and 5-vector notation, where $e(q) = \{e_i(q), i = 0, 1, 2, 3, 4\}$ and

$$e_0(q) = 1 \qquad e_1(q) = \omega(q) = 1 - \tfrac{1}{2}c_1 - \tfrac{1}{2}c_2$$
$$e_2(q) = \tfrac{1}{2}c_1 - \tfrac{1}{2}c_2 \qquad e_3(q) = s_1 \qquad e_4(q) = s_2. \tag{3.12}$$

In this basis we can write

$$\delta L_{qq'} = e(q)Ve(q')C_{qq'} = \sum_{i,j} e_i(q)V_{ij}e_j(q')C_{qq'}$$
$$\Lambda_{qq'} = e(q)\bar{V}e(q')C_{qq'} \tag{3.13}$$

$$V = b \begin{pmatrix} \begin{array}{cc|c} 0 & 0 & \\ -1 & 1 & 0 \\ \hline & & 1 \\ 0 & & \frac{1}{2} \\ & & & \frac{1}{2} \end{array} \end{pmatrix} \qquad \bar{V} = b \begin{pmatrix} \begin{array}{cc|c} -z & -1 & \\ -1 & 1 & 0 \\ \hline & & 1 \\ 0 & & \frac{1}{2} \\ & & & \frac{1}{2} \end{array} \end{pmatrix}. \quad (3.14)$$

Verification of (3.13) is straightforward by working out the matrix multiplications using (3.12), (3.11) and (3.6). Regarding notation we follow in general the convention that 5×5 matrices are denoted by upper case letters and 5-vectors by lower case letters.

We consider a typical term in the perturbation expansion (3.10) and insert for $\delta L_{qq'}$ and $\Lambda_{qq'}$ the expressions (3.13), namely

$$\delta L_{qq'} = N^{-1} \sum_n \exp[i(q - q')n] c_n e(q) V e(q') \quad (3.15)$$

and a similar expression for $\Lambda_{qq'}$ with V replaced by $\bar{V}$. The summations run in fact over impurity sites only. This yields

$$((\delta Lg)^l \Lambda)_{qq'} = \sum_{q_1 \cdots q_l} \delta L_{qq_1} g(q_1) \delta L_{q_1 q_2} \cdots g(q_l) \Lambda_{q_l q'}$$

$$= N^{-1} \sum_{n_1 n_2 \ldots n_l m} c_{n_1} c_{n_2} \ldots c_{n_l} c_m \exp(iqn_1) e(q) V G(n_1 - n_2) V G(n_2 - n_3) \ldots$$

$$\times V G(n_l - m) \bar{V} e(q') \exp(-iq'm). \quad (3.16)$$

Here we have introduced the 5×5 matrix $G(n, z)$ with elements

$$G_{ij}(n, z) = \int_q \exp(-inq) e_i(q) e_j(q) / [z + \omega(q)] \quad (3.17)$$

and we used the shorthand notation

$$\int_q (\ldots) = N^{-1} \sum_{q \in \text{1BZ}} (\ldots) \underset{N \to \infty}{\simeq} (2\pi)^{-2} \int_{-\pi}^{\pi} \int dq_x \, dq_y (\ldots). \quad (3.18)$$

In the thermodynamic limit ($N \to \infty$) the q summation is replaced by an integral over the first Brillouin zone. A matrix element of the lattice Green function in (3.17) is essentially the (Laplace transformed) probability for a displacement n on a uniform lattice, and the factors V and $\bar{V}$ represent 'collisions' of the RW with an impurity.

Suppose that two successive site labels, n' and n'', refer to the same site, then $c_{n'} c_{n''} = (c_{n'})^2$ and $G(n' - n'', z) = G(0, z)$ is essentially the probability of return to the same site on a uniform lattice. If we sum all possible returns to the same impurity site n ('repeated ring collisions' in kinetic theory language) we obtain the single impurity T and $\bar{T}$ matrix:

$$T(z) = \sum_{l=0}^{\infty} (V G(0))^l V = (1 - V G(0))^{-1} V$$

$$\bar{T}(z) = \sum_{l=0}^{\infty} (V G(0))^l \bar{V} = (1 - V G(0))^{-1} \bar{V}. \quad (3.19)$$

The properties of these matrices, needed in later sections, are studied in appendix 1.

By carrying out the T matrix resummation in (3.16) we obtain

$$g^{-2}h_m = N \sum_{l=0} ((\delta L_m g)^l \Lambda_m)_{qq}$$

$$= mce(q)\bar{T}e(q) + \sum_{l=1}^{\infty} \sum_{n_1 \ldots n_l}' c_{n_1} \ldots c_{n_l} c_0 \exp(-iqn_1)$$

$$\times e(q)R(n_1 - n_2)R(n_2 - n_3) \ldots R(n_l)\bar{T}e(q) \qquad (3.20)$$

where c_n is only non-vanishing for m specified sites, one of which is chosen to be the origin.

The prime on the summation sign indicates the constraint that any pair of consecutive site labels be different. We have further introduced

$$R(n, z) = T(z)G(n, z) \qquad (3.21)$$

We first consider the T matrix expansion (3.20) for $m = 1$. Since the lattice sum contains at least two impurities (at $n = 0$ and at $n_l \neq 0$) only the first term survives and we find for the coefficient f_1 in (3.5) the exact result

$$f_1(q, z) = h_1(q, z) = g^2(q)e(q)\bar{T}e(q) \qquad (3.22)$$

where $\bar{T}(z)$ sums all possible returns of the RW to a single impurity. Of course, f_0 in (3.5) equals $g(q)$.

Next, we consider $f_2(q, z)$ in (3.5) which requires $h_2(n) - 2h_1$, describing the response function of a system with, say, impurity '1' at the origin and impurity '2' at $n \neq 0$, so that $c_m = \delta_{m0} + \delta_{mn}$. Since the constraints of the T matrix expansion requires that consecutive site labels in the sequence $c_{n_1}c_{n_2} \ldots c_0$ be different, the only terms present in (3.20) describe sequences [12], [121], [1212], [12121], ... We separately sum the subsequences of all walks starting and ending at the *same* site, yielding

$$(h_2(n) - 2h_1)_{same} = e(q)[1 - R(n)R(-n)]^{-1}R(n)R(-n)T\bar{e}(q)$$

$$= e(q)[1 - \hat{R}^2(n)]^{-1}\hat{R}^2(n)\bar{T}e(q).$$

We have used the relation $R(-n) = SR(n)S$ due to symmetry under inversion (see (A2.2) of appendix 2) and introduced $\hat{R}(n, z) = R(n, z)S$, where S is a diagonal matrix (see (A2.1)) with elements $S = \text{diag}\{1, 1, 1, -1, -1\}$. The sum of all walks, starting at the first impurity and ending at the second one, can be summed similarly. Therefore, our final result for the coefficient $f_2(q, z)$ of the response function in (3.5) is

$$f_2(q, z) = \tfrac{1}{2}g^2(q)e(q) \sum_{n \neq 0} (1 - \hat{R}^2(n))^{-1}\hat{R}^2(n)\bar{T}e(q)$$

$$+ \tfrac{1}{2}g^2(q)e(q) \sum_{n \neq 0} \exp(iqn)(1 - \hat{R}^2(n))^{-1}\hat{R}(n)\bar{T}Se(q). \qquad (3.23)$$

From these results in combination with (3.1) we can calculate all moments of displacement, return probabilities, etc, exact to $O(c^2)$ terms included.

4. Frequency-dependent transport coefficients

In this section we derive a general expression for the Laplace transform $\Phi(z)$ of the VACF $\varphi(t)$, defined in (2.4), and for the 'frequency-dependent' conductivity $\Xi(z) = \langle \psi \rangle \Phi(z)$, valid for all z values and exact to $O(c^2)$ terms included. The limiting values of these quantities as $z \to 0$ determine the static transport properties.

The VACF $\Phi(z)$ can be extracted from the response function $F(q, z)$ using (2.4), (2.11) and (2.14), namely $\Phi(z) = -\frac{1}{2}z^2 F''(0, z)$, where we use the notation $A'(0, z) \equiv \partial A(q, z)/\partial q_x|_{q=0}$, etc. Since $\Xi(z) = \langle \psi \rangle \Phi(z)$ it follows from (3.1) that

$$\Xi(z) = -\frac{1}{2}z^2 \langle H''(0, z) \rangle. \tag{4.1}$$

The calculation of $\Xi(z)$ to zeroth and first order in c is simple and yields $\Xi(z) = \frac{1}{4} - ct_3(z) + O(c^2)$. This follows from (3.1) with $f_0(q, z) = g(q)$ and $f_1(q, z)$ in (3.22), in combination with (3.12) and (A1.5).

The calculation of the $O(c^2)$ term, $-\frac{1}{2}z^2 f_2''(0, z)$, as defined in (3.23) is much more involved and we only sketch some intermediate steps. First, observe that the 5-vectors $e(q)$ in (3.23) and their q_x derivatives at $q = 0$ are given by

$$e_j(0) = \delta_{j0} \qquad e_j'(0) = \delta_{j3} \qquad e_j''(0) = \frac{1}{2}(\delta_{j1} - \delta_{j2}).$$

Next, we differentiate (3.23) twice with respect to q_x and use the relation $e(0)\hat{R}(n, z) = e(0)R(n, z)S = 0$, as follows from (3.21) and $e(0)T(z) = 0$ (see (A1.1)). The result reads in a schematic notation

$$-\frac{1}{2}z^2 f_2''(0, z) = -\frac{1}{2}e'(0)(A + B)e'(0) - \frac{1}{4}e''(0)(A + B)e(0) - \frac{1}{2}e'(0)in_x Be(0) \tag{4.2}$$

where we imply summations over n and use the abbreviations

$$\begin{aligned} A &= (1 - \hat{R}^2(n))^{-1}\hat{K}^2(n)\bar{T} \\ B &= (1 - \hat{R}^2(n))^{-1}\hat{R}(n)\bar{T}S. \end{aligned} \tag{4.3}$$

In the first term on the RHS of (4.2) a common factor $(1 - \hat{R}(n))$ in the numerator and denominator cancels and $\bar{T}(z)$ may be replaced by $t(z)$ (compare (A1.4) and (A1.5)) since only the (33) element enters. The first term contains the only n summation that extends over the entire lattice. All remaining terms in (4.2) are restricted to the four NN sites ρ on account of the relation $\hat{R}(n)\bar{T}e(0) = -bt\alpha(\rho)\delta_{n\rho}$ (see (A1.8)). In the second term on the RHS of (4.2) we cancel a common factor $(1 + \hat{R}(n))$ in the numerator and denominator. The resulting expression has the structure of a 5-vector $w(\rho)$ with components $\{w(\rho)\}_i$. Use of the cubic symmetry shows that the component with $i = 2$ disappears from the NN sum and that we can express everything in the NN vector $\rho_1 = \{1, 0\}$.

The last term in (4.2) is again restricted to NN sites ρ, where only the x components of $\rho_1 = \{1, 0\}$ and $\rho_3 = \{-1, 0\}$ are non-vanishing. Their contributions are equal by cubic symmetry. Combining the above results yields then

$$\begin{aligned} -\frac{1}{2}z^2 f_2''(0, z) = \sum_{n \neq 0} [(1 + \hat{R}(n))^{-1}\hat{R}(n)t]_{33} + ib\{[(1 + R(\rho_1))^{-1} \\ + (1 - \hat{R}(\rho_1))^{-1}]t\alpha(\rho_1)\}_3 + b[(1 - \hat{R}(\rho_1))^{-1}t\alpha(\rho_1)]_1. \end{aligned} \tag{4.4}$$

The purpose of the following steps is to transform the $O(c^2)$ terms in (4.4) by making the matrix elements real and to simplify them further by dividing numerators and denominators by a common factor $t(z)$. We start by introducing the real matrix

$$g(n, z) = S_c \int g(q, z) \exp(-iqn)\tilde{e}(q)e(q)S_c \tag{4.5}$$

where the 5-vectors $\tilde{e}(q)$ and $e(q)$ have been defined in (A1.7) and (3.12), respectively. The transformation matrix S_c is diagonal with matrix elements $S_c = \text{diag}\{1, 1, 1, i, i\}$ with $i = \sqrt{-1}$, and has the properties $S_c^+ S_c = 1$ and $S_c^2 = S$, where S is defined in (A2.1).

To prove that (4.5) is real one takes the complex conjugate and uses the third symmetry relation of (A2.2). Next we observe that

$$\hat{R}(n, z) = t(z) \int g(q, z) \exp(-iqn)\tilde{e}(q)e(q)S$$

$$= S_c^+ t(z)g(n, z)S_c. \tag{4.6}$$

Here we have used $\hat{R}(n) = R(n)S$ and (3.21), (3.17) and (A1.6) and the properties of S_c. By inserting (4.6) into the first term of (4.4) we find

$$[(1 + \hat{R}(n))^{-1}\hat{R}(n)t]_{33} = [(t^{-1} + g(n))^{-1}g(n)t]_{33} \equiv k_{33}(n, z). \tag{4.7}$$

We have also used the relation $M_{33} = \{S_c^+ M S_c\}_{33}$, valid for the (33) element of a matrix M. Similarly we find for the next term in (A3.3)

$$ib[(1 + \hat{R}(\rho_1))^{-1}t\alpha(\rho_1)]_{33} = b[(t^{-1} + g(\rho_1))^{-1}a(\rho_1)]_3 \equiv Q_3(z) \tag{4.8}$$

where we have defined $a(\rho_1) = S_c\alpha(\rho_1) = \frac{1}{4}\{0, -1, 1, 2, 0\}$. Finally we introduce

$$m(z) \equiv S_c b(1 - \hat{R}(\rho_1))^{-1}t\alpha(\rho_1)$$

$$= b(t^{-1} - g(\rho_1))^{-1}a(\rho_1). \tag{4.9}$$

After these transformations and definitions we have

$$-\tfrac{1}{2}z^2 f''(0, z) = \sum_{n \neq 0} k_{33}(n, z) + Q_3(z) + m_3(z) + m_1(z). \tag{4.10}$$

The final simplification is a proof of the identity $m_1(z) + m_3(z) = 0$. In fact we shall prove the more general result that the system of linear equations

$$(t^{-1} - g(\rho_1))m = b\alpha(\rho_1) \tag{4.11}$$

with $g(\rho_1) \equiv g(\rho_1, z)$ has the exact solution $m(z) = \frac{1}{4}b^2\{0, -1, 1, 1, 0\}$, which implies the above identity. To verify the solution we first note that $g_{0i}(n, z) = 0$ on account of (4.5) and (A1.7). Furthermore $g_{4j}(\rho_1, z) = 0$ for $j \neq 4$, as can be seen from (A3.4) because the integrand for $n = \rho_j$ is an odd function of q_y. Furthermore $a_0 = a_4 = 0$, so that $m_0(z) = m_4(z) = 0$. Hence we can restrict ourselves to the (123) subspace.

The simplest way to proceed is to substitute the solution into (4.11) and show that the resulting equations are identities. This ends the proof of the identity.

Inserting these simplifications into (4.4) and combining this with the low density results, derived below (4.1), we obtain the following result for the frequency-dependent conductivity $\Xi(z) = \langle \psi \rangle \Phi(z)$ and VACF $\Phi(z)$, where $\langle \psi \rangle = 1 - bc$, exact to $O(c^2)$ terms included:

$$\Xi(z) = \tfrac{1}{4} - ct_3(z) + c^2(K_{33}(z) + Q_3(z)) + O(c^3)$$

$$\Phi(z) = \tfrac{1}{4} + \tfrac{1}{4}cb(1 + bc) - c(1 + bc)t_3(z) + c^2(K_{33}(z) + Q_3(z)) + O(c^3) \tag{4.12}$$

where t_3, K_{33} and Q_3 are elements of 5×5 matrices t and K and a 5-vector Q, defined as

$$K(z) = \sum_{n \neq 0} k(n, z) = \sum_{n \neq 0} (t^{-1}(z) + g(n, z))^{-1}g(n, z)t(z)$$

$$Q(z) = (t^{-1}(z) + g(\rho_1, z))^{-1}a(\rho_1)b \tag{4.13}$$

where $\rho_1 = \{1, 0\}$ is a NN site of the origin, and the column 5-vector $a(\rho_1)$ has the elements $a(\rho_1) = \{0, -\frac{1}{4}, \frac{1}{4}, \frac{1}{2}, 0\}$.

The lattice Green function $g(n, z)$ is a transformed version of (3.17) and is defined in (4.5). The diagonal $t(z)$ matrix is defined in (A1.4) and (A1.2). The general expressions (4.12) for the VACF and the frequency-dependent conductivity $\Xi(z)$ are the main results of this paper. They were the starting points for the calculations reported in I.

To obtain explicit values for diffusion coefficient, conductivity and VACF at different σ values one has to calculate lattice Green functions $g(n, z)$ for all sites, to invert the matrices $(t^{-1}(z) + g(n, z))$ and to carry out the lattice summations in (4.13). This still requires a great deal of further analysis and numerical calculations, which will be given in a subsequent article.

For the special case of percolation (see I), where the impurity sites are excluded sites with $\sigma = 0$ or $b = 1$, we have already evaluated the $O(c)$ and $O(c^2)$ coefficients in the density expansion of the diffusion coefficient and the DC conductivity. Our results are in very good agreement with the results obtained by Harris and Kirkpatrick (1977) from computer simulations, and with the approximate analytic result of Watson and Leath (1974) for the DC conductivity, $\Sigma(c) = \frac{1}{4}(1 - \pi c + \frac{1}{2}\pi c^2)$.

The VACF in the percolation case has also been calculated in I and our detailed results are in very good agreement with the recent results of computer simulations by Frenkel (1987) on this hopping model.

Acknowledgments

Appendix 1

This appendix deals with properties of the T matrix. Consider first the matrix $G(0, z)$ defined in (3.17). From the parity of the integrand under the reflections $q_x \leftrightarrow -q_x$, and $q_y \leftrightarrow -q_y$ and $q_x \leftrightarrow q_y$ it follows that $G(0, z)$ has the same structure as the matrix $\bar{V}$ in (3.14), namely it is diagonal except in the (01) subspace. To calculate $T(z)$ and $\bar{T}(z)$ we only have to invert a 2×2 matrix. This gives

$$T(z) = \begin{pmatrix} \begin{array}{cc|ccc} 0 & 0 & & & \\ -t_1 & t_1 & & 0 & \\ \hline & & t_2 & & \\ 0 & & & t_3 & \\ & & & & t_4 \end{array} \end{pmatrix} \tag{A1.1}$$

with

$$1/t_1(z) = 1/b - g_{11}(0, z)$$

$$1/t_2(z) = 1/b - g_{22}(0, z) \tag{A1.2}$$

$$1/t_3(z) = 1/t_4(z) = 2/b + g_{33}(0, z).$$

Here we have introduced the matrix $g(n, z)$ (see (4.4)) with

$$g_{11}(0, z) = \int g(e_1 - 1)e_1 = G_{11}(0, z) - G_{10}(0, z)$$

$$g_{22}(0, z) = \int ge_2^2 = G_{22}(0, z) \qquad (A1.3)$$

$$g_{33}(0, z) = -\int ge_3^2 = -G_{33}(0, z) = -G_{44}(0, z)$$

where the integration symbol denotes the average (3.19) over the ıBZ with $g = g(q, z) = (z + \omega(q))^{-1} = (ze_0 + e_1)^{-1}$.

In the body of the paper we also use the closely related diagonal t matrix, labelled by $i, j \in \{0, 1, 2, 3, 4\}$ with elements

$$t(z) = \mathrm{diag}\{1, t_1, t_2, t_3, t_3\}. \qquad (A1.4)$$

In a similar manner we find for $\bar{T}(z)$ in (3.19)

$$\bar{T}(z) = \begin{pmatrix} \begin{array}{cc|cc} -zb & -b & & \\ -b & \bar{t}_1 & & 0 \\ \hline & & t_2 & \\ & 0 & & t_3 \\ & & & & t_3 \end{array} \end{pmatrix} \qquad (A1.5)$$

where $\bar{t}_1(z) = t_1(z)[1 - b \int g(e_1 - 1)e_0]$. To prove the relation $\bar{T}_{10}(z) = -b$ we used $\int g(e_1 - 1)(e_1 + ze_0) = \int (e_1(q) - 1) = 0$ as follows from the definition of g and $ze_0 + e_1 = z + \omega(q)$.

Next we summarise some properties of T matrices, needed in the body of the paper:

$$T(z)e(q) = t(z)\tilde{e}(q) \qquad (A1.6)$$

as follows from the previous equations, where $\tilde{e}(q)$ is a 5-vector with elements

$$\tilde{e}(q) = \{0, e_1 - 1, e_2, e_3, e_4\} \qquad (A1.7)$$

with $e_j(q)$ in (3.12). A further property used is

$$\hat{R}(n, z)\bar{T}(z)e(0) = -bt(z)\alpha(\rho)\delta_{n\rho} \qquad (A1.8)$$

where the occurring symbols are defined in (3.21), (3.19) and above (3.23). The ʀʜs is only non-vanishing if n is a ɴɴ site ρ_i of the origin, where $\rho_{1,3} = \{\pm 1, 0\}$ and $\rho_{2,4} = \{0, \pm 1\}$. Here the 5-vector $\alpha(\rho_1)$ is given by

$$\alpha(\rho_1) = \tfrac{1}{4}\{0, -1, 1, -2i, 0\} \qquad (A1.9)$$

with $i = \sqrt{-1}$. The remaining $\alpha(\rho_i)$ can be obtained through the symmetry relations of appendix 2.

The remaining part of this appendix is a derivation of (A1.8). We first observe that $e(0) = \{1, 0, 0, 0, 0\}$ so that $\bar{T}(z)e(0) = \bar{T}_{.1}(z)$ represents the first column vector of $\bar{T}(z)$ with elements $\bar{T}_{.1}(z) = \{-bz, -b, 0, 0, 0\}$. Consequently the components of the 5-vector in (A1.8) are

$$(\hat{R}(n, z)\bar{T}(z)e(0))_j = -b(\hat{R}_{j1}(n, z) + z\hat{R}_{j0}(n, z)). \qquad (A1.10)$$

Using the definitions (3.19) and (4.2) we can express $R(n, z)$ as

$$\hat{R}(n, z) = T \int gee \exp(-iqn)S = t \int g\tilde{e}e \exp(-iqn)S.$$

Consequently the RHS of (A1.10) can be written as

$$\text{RHS(A1.12)} = -bt_j(z) \int g\tilde{e}_j(ze_0 + e_1) \exp(-iqn) = -bt_j(z)\alpha_j(\rho)\delta_{n\rho}$$

where we have used the relation $g^{-1} = ze_0 + e_1$ and defined $\alpha(\rho) = \int \tilde{e} \exp(-iq\rho)$. Combining these equations yields finally (A1.8). The first equality follows from (A1.3) and shows that the LHS is independent of z, apart from the factor $t_j(z)$. Hence the second equality.

The angular average $\int_q \dots$ is only non-vanishing if $n = \rho$, since the 5-vector $\tilde{e}(q)$, defined in (A1.7), is a linear combination of c_1, c_2, s_1 and s_2 (see (3.12)). For $\rho_1 = \{1, 0\}$ the average yields the result (A1.9) and combination of the above equations proves (A1.8).

Appendix 2

This appendix deals with symmetry properties under reflections of the lattice Green functions $G(n, z)$ in (3.17) and $g(n, z)$ in (4.5) and related 5×5 matrices and 5-vectors. We define the following transformation matrices, whose square equals the unit matrix:

$$S_x = \begin{pmatrix} 1 & & & & \\ & 1 & & & \\ & & 1 & & \\ \hline & & & -1 & \\ & & & & 1 \end{pmatrix} \qquad S_y = \begin{pmatrix} 1 & & & & \\ & 1 & & & \\ & & 1 & & \\ \hline & & & 1 & \\ & & & & -1 \end{pmatrix}$$

$$S = S_xS_y = \begin{pmatrix} 1 & & & & \\ & 1 & & & \\ & & 1 & & \\ \hline & & & -1 & \\ & & & & -1 \end{pmatrix} \qquad S_{xy} = \begin{pmatrix} 1 & & & & \\ & 1 & & & \\ & & 1 & & \\ \hline & & & 0 & 1 \\ & & & 1 & 0 \end{pmatrix}.$$

$$(A2.1)$$

They correspond respectively to reflections with respect to the y axis, the x axis, the origin and the diagonal $n_x = n_y$. One can easily verify that matrix $G(n, z)$ in (3.17) satisfies the following symmetry relations under reflections:

Reflection $n \to n'$	Symmetry relations
$\{n_x, n_y\} \to \{-n_x, n_y\}$	$G(n', z) = S_xG(n, z)S_x$
$\{n_x, n_y\} \to \{n_x, -n_y\}$	$G(n', z) = S_yG(n, z)S_y$
$\{n_x, n_y\} \to \{-n_x, -n_y\}$	$G(-n, z) = SG(n, z)S$
$\{n_x, n_y\} \to \{n_y, n_x\}$	$G(n', z) = S_{xy}G(n, z)S_{xy}.$

$$(A2.2)$$

The matrices V, $\bar{V}$, T, $\bar{T}$ and $G(0, z)$ in (3.14), (3.19), (A1.1) and (A1.3) are invariant under reflections, namely they commute with all S matrices in (A2.1). Consequently

$\hat{R}(n, z)$ in (3.21), $g(n, z)$ in (4.5), and products of these matrices satisfy the same symmetry relations (A2.2).

The transformation properties of a 5-vector $\alpha(n)$ (such as in (4.4) and (A1.9)) are similar, namely

$$\alpha(n') = S\alpha(n) \qquad \text{for } n \to n' \tag{A2.3}$$

where n' and S' stand for any of the transformations in (A2.2).

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