Systematic density expansion for random resistor networks

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Abstract. We present a systematic density expansion for calculating transport properties, return probabilities and spectral densities for lattice hopping models with diluted randomness. The method is applied to a disordered square lattice where the bonds carry conductances σ_1 and σ_2 in concentrations c and $1-c$, respectively, and we calculate explicitly the $O(c)$ and $O(c^2)$ contributions to the static conductivity (diffusion coefficient) and to the coefficient $\beta(c)$ of the long time tail in the velocity autocorrelation function (VACF) $\langle v_x(0)v_x(t)\rangle \simeq -\beta(c)t^{-2}$.

To $O(c)$ the VACF is calculated for all times and shows negative correlations (cage effect) for all times.

Diffusive systems with static disorder, such as Lorentz gases, dynamic percolation, ants in a labyrinth, termite problems, random resistor networks, networks with normal and superelastic springs, etc, have received much attention recently. Such problems can be modelled by random walks on disordered lattices or equivalently by lattice dynamics of disordered harmonic crystals.

For the case of diluted randomness, where only a random fraction c of bonds or sites of the host lattice have been replaced by impurities, we present a systematic expansion of DC and AC transport properties in powers of the impurity concentration c. In particular, we study a random walk on a square lattice with two types of bonds: impurity bonds with a transition rate ('conductance') σ and bonds of the host lattice with conductance $\sigma_0 = 1$. The model is a special case of the random barrier model (Haus *et al* 1983, Derrida 1983), and for the 1D chain many properties have been calculated exactly by Denteneer and Ernst (1984); for instance, the static diffusion coefficient is given by $D^{-1} = 2[1 + c(1-\sigma)/\sigma]$. The model describes bond percolation for $\sigma = 0$ and the termite problem for $\sigma \gg 1$ (Hong *et al* 1986).

The purpose of this paper is to outline a kinetic theory approach for calculating static and time-dependent properties of RW on disordered lattices and to illustrate the power of this method by showing for the 2D square lattice exact low density results for different quantities such as the static and time-dependent diffusion coefficient, the velocity autocorrelation function and frequency-dependent conductivity. The method can also be used for different dimensionality for higher moments of displacements and Burnett coefficients, probabilities of return and first passage, etc.

Since calculations are lengthy, as is usual in kinetic theory, we have left out almost all analytical derivations of equations, as well as discussions on the methods used in

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the numerical evaluation of the occurring lattice sums. These details together with further applications will be published elsewhere.

Most theoretical and computer studies have concentrated on percolation problems (ants and termites) in the close vicinity of the percolation threshold. The standard methods to calculate transport properties for general impurity concentration are effective medium type approximations (EMA) (Kirkpatrick 1973, Watson and Leath 1974, Odagaki 1986, Garboczi and Thorpe 1986), single impurity approximations (Harris and Kirkpatrick 1977, Roerdink and Shuler 1985), 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). Systematic density expansions for dynamics quantities (Fish and Harris 1978) are not so well developed as for static ones (Coniglio *et al* 1977, Gaunt and Sykes 1983). Here we calculate the static conductivity or the static diffusion coefficient $D(c, \sigma)$ and the coefficient of the long time tail of the VACF, $\phi(t) \simeq -\beta(c, \sigma)/t^2$, exact to terms of $O(c^2)$ included, for several values of the conductance ratio σ.

We further calculate the complete time dependence of the VACF to $O(c)$ and explain the cage effect (existence of negative velocity correlations) in terms of repeated backscattering by impurities ('repeated ring collisions'). Our detailed predictions of the intermediate and long time behaviour of the VACF are presumably sufficient to allow a detailed comparison with the results of computer simulations, as appeared possible for the corresponding site problem (Nieuwenhuizen *et al* 1986, Frenkel 1986). For deterministic Lorentz models a quantitative agreement on long time tails in the VACF, obtained from computer simulations and kinetic theory, is still lacking (Alder and Alley 1983, Ernst *et al* 1984). In dense fluids, on the other hand, the agreement is satisfactory (Erpenbeck and Wood 1985).

The quantities to be explicitly considered are the mean square displacement and the related time-dependent diffusion coefficient:

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

where $\tilde{D}(\infty) = D$ is the static diffusion coefficient and $\phi(t)$ is the lattice analogue of the velocity autocorrelation function (VACF) $\langle v_x(0)v_x(t)\rangle$.

Our system is a square lattice with unit lattice distance, with N sites labelled $n = (n_x, n_y)$, and with periodic boundary conditions. The lattice has two types of bonds: impurity bonds with 'conductance' σ (occurring randomly with concentration c) and bonds with $\sigma_0 = 1$ (their concentration is $(1-c)$). On this random lattice we consider a random walker (RW), whose probability distribution is described by the master equation:

$$\dot{p}_n = \tfrac{1}{4}\sum_\rho \psi_{n,n+\rho}(p_{n+\rho} - p_\rho) \equiv -(Lp)_n \tag{2}$$

where ρ denotes a nearest-neighbour lattice vector: $\pm\hat{e}_x, \pm\hat{e}_y$. To every bond $(n, n+\rho)$ we have assigned a random variable $\psi_{n,n+\rho} = 1 - bc_{n,n+\rho}$, where $c_{n,n+\rho} = 1$ with probability c and $c_{n,n+\rho} = 0$ with probability $(1-c)$. Thus, the impurity bonds ($c_{n,n+\rho} = 1$) have a conductance $\sigma = 1 - b \geq 0$, and those of the host lattice ($c_{n,n+\rho} = 0$) have $\sigma_0 = 1$. The model considered here is a continuous time RW (namely with a Poissonian distribution of pausing times) and it describes unbiased or blind ants ($\sigma = 0$) or termites ($\sigma \gg 1$). In a time $\mathrm{d}t$ the probability for hopping across a bond $(n, n+\rho)$ is $(\psi_{n,n+\rho}\,\mathrm{d}t/4)$, which

equals $(\sigma \, \mathrm{d}t/4) = [(1-b) \, \mathrm{d}t/4]$ for an impurity bond and $(\mathrm{d}t/4)$ for a bond of the host lattice. The probability for the RW to remain on the same site in a time $\mathrm{d}t$ is $(1 - \Sigma_\rho \, \psi_{n,n+\rho} \, \mathrm{d}t)$, which is by definition close to unity (also for termites). Once a termite hits a 'superconducting' bond or cluster of superconducting bonds it will leave the superconducting bonds on the average only in 1 out of σ hops. The termites considered here are somewhat different from the 'Boston' and 'Tel Aviv' termites (Hong *et al* 1986).

The main quantity of interest is the average probability distribution for a displacement from site m to site n in a time t, defined as $P_{n-m}(t) = N^{-1} \langle p(nt|m0) \rangle$, where $p(nt|m0) = [\exp(-tL)]_{nm}$ is the solution of the master equation with initial condition $p(n0|m0) = \delta_{nm}$. The average is taken over the probability distribution of the set of random variables $\{c_{n,n+\rho}\}$.

The following *symmetries* hold for $P_n(t)$ (Straley 1977). Since (2) is invariant under the mapping $\sigma \to \sigma' = 1/\sigma$, $t \to t' = \sigma t$ and $c \to c' = 1 - c$ where c refers to the random variables $\{c_{n,n+\rho}\}$ and to their average $\langle c_{n,n+\rho} \rangle = c$ we have

$$P_n(t, c, \sigma) = P_n(\sigma t, 1-c, 1/\sigma). \tag{3}$$

This implies $D(c, \sigma) = \sigma D(1 - c, 1/\sigma)$ for the static diffusion coefficient and $\phi(t, c, \sigma) = \sigma^2 \phi(\sigma t, 1-c, 1/\sigma)$ for the VACF. Since a square lattice with a random mixture of two different conductances is self-dual we further have $D(c, \sigma) D(c, 1/\sigma) = D_0^2$ where $D_0 = D(0, \sigma) = \frac{1}{4}$ is the diffusion coefficient of the host lattice. Combination of both symmetries yields $D(c, \sigma) D(1 - c, \sigma) = \sigma D_0^2$, so that for the half-filled lattice $D(\frac{1}{2}, \sigma) = \sqrt{\sigma} D_0$. Self-duality induces an additional symmetry relation in the coefficient of the long time tail, as will be discussed later on.

A *resolvent formalism* is introduced for the response function $F(q, z)$, which is the Fourier-Laplace transform of $P_n(t)$, namely

$$F(q, z) = N^{-1} \sum_{n,m} \exp[iq(n-m)] \langle (z + \hat{L})^{-1} \rangle_{nm} = \langle (z + L)^{-1} \rangle_{qq} \tag{4}$$

where we have defined the Fourier representation $A_{qq'}$ of the matrix $\hat{A}_{nm}$ in coordinate representation $A_{qq'} = N^{-1} \Sigma_{nm} \exp[iq(n-m)] \hat{A}_{nm}$. The response function generates the moments of displacement, so that the Laplace transformed mean square displacement becomes

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

Next we split L into $L = L^0 - \delta L$, where L^0 and $-\delta L$ are obtained from (2) by replacing $\psi_{n,n+\rho}$ respectively by 1 and $-b c_{n,n+\rho}$. The Fourier transforms of these matrices are

$$L_{qq'}^0 = \omega(q) \delta_{qq'}$$

$$\delta L_{qq'} = (b/4N) \sum_{n\alpha} c_n^\alpha \exp[i(q-q')n] \varepsilon_\alpha^*(q) \varepsilon_\alpha(q)$$

where $\varepsilon_\alpha(q) = 1 - \exp(-iq_\alpha)$ with $\alpha = x, y$ and $\omega(q) = 1 - \frac{1}{2} \cos q_x - \frac{1}{2} \cos q_y$ and $c_n^\alpha = c_{n,n+\rho_\alpha}$ is used as a more compact way of bond labelling. Note that only impurity bonds contribute to δL. We proceed with the perturbation expansion of $\langle (z + L^0 - \delta L)^{-1} \rangle$ in powers of δL, yielding $F(q, z) = g(q) + g^2(q) M(q, z)$, where $M(q, z) = \Sigma_{l=0} \langle (\delta L g)^{l-1} \delta L \rangle_{qq}$ with $g_{qq'} = g(q) \delta_{qq'}$ and $g(q) = (z + \omega(q))^{-1}$.

Using a *T-matrix resummation* we regroup the terms in the perturbation expansion by adding successive terms referring to the same impurity, where the single impurity

T matrix is

$$T(z) = \tfrac{1}{4}b[1 - bJ(z)]^{-1}$$

$$J(z) = \tfrac{1}{2} \int_q \omega(q)[z + \omega(q)]^{-1}. \tag{5}$$

Here $\int_q \ldots$ represents a sum (cq integral if $N \to \infty$) over the first Brillouin zone (1BZ), namely $\int_q \ldots = N^{-1} \Sigma_{q \in 1BZ} \ldots = (2\pi)^{-2} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} dq_x \, dq_y \ldots$. The following T-matrix expansion is obtained:

$$M(q, z) = N^{-1} \sum_{n\alpha} \langle c_n^\alpha \rangle \varepsilon_\alpha \varepsilon_\alpha^* T + N^{-1} \sum_{n\alpha, m\beta}' \langle c_n^\alpha c_m^\beta \rangle \exp[iq(n-m)] \varepsilon_\beta \varepsilon_\alpha^* T G_{\alpha\beta}(n-m) T$$

$$+ N^{-1} \sum_{n\alpha, m\beta, s\gamma}' \langle c_n^\alpha c_m^\beta c_s^\gamma \rangle \exp[iq(n-s)] \varepsilon_\gamma \varepsilon_\alpha^* T G_{\alpha\beta}(n-m)$$

$$\times T G_{\beta\gamma}(m-s) T + \ldots \tag{6}$$

where $\varepsilon_\alpha = \varepsilon_\alpha(q)$ and where the prime on the summation signs indicates that consecutive bond labels are different, namely $(n\alpha) \neq (m\beta)$, $(m\beta) \neq (s\gamma) \ldots$. We have further introduced

$$G_{\alpha\beta}(n, z) = \int_q \exp(-iqn)\varepsilon_\alpha(q)\varepsilon_\beta^*(q)/[z + \omega(q)] \tag{7}$$

which is essentially (apart from goniometric factors $\varepsilon_\alpha \varepsilon_\beta^*$ the probability for a displacement n on a *uniform* lattice.

Next, the *density expansion* is considered. The first term in (6) accounts for all possible visits of the RW to a single impurity ('repeated ring collisions with a single scatterer') and reduces to $4c\omega(q)T(z)$. This is the exact result to linear order in the impurity concentration, since all higher-order terms in (6) involve at least two scatterers. To obtain the $O(c^2)$ contributions we select from (6) the collision sequences involving exactly two impurities. They have the structure [12], [121], [1212], [12121], [121212] .... We further observe that the corresponding density fluctuation reduces to $\langle c_1 c_2 c_1 c_2 c_1 \ldots \rangle = \langle c_1 c_2 \rangle = c^2$. By summing all possible walks between two different impurities we finally obtain an expression for $F(q, z)$, exact to $O(c^2)$ terms included. From the generating function $F(q, z)$ one can extract the Laplace transform of the VACF, denoted by $\Phi(z) = \tfrac{1}{2}z^2 \langle n_x^2 \rangle(z)$, and one obtains after some algebra the VACF, exact to $O(c^2)$ terms included

$$\Phi(z) = \tfrac{1}{4} - cT(z) - c^2 T(z) \left(\sum_{n \neq 0} R_{xx}(n, z)/(1 - R_{xx}^2(n, z)) \right.$$

$$\left. + \sum_{n\beta}' R_{x\beta}^2(n, z)/(1 - R_{x\beta}^2(n, z)) \right) + O(c^3). \tag{8}$$

The prime on the sums indicates that the terms with bond label $(n\beta) = (0x)$ are excluded (restriction from the T-matrix resummation). The integral $R_{\alpha\beta}(n, z) = G_{\alpha\beta}(n, z)T(z)$ contains a single crossing from scatterer 1 to scatterer 2 and all possible returns to scatterer 2, where n is the difference in positions between scatterer 2 and 1. We further note that the dependence on the ratio of the two conductances ($\sigma \equiv 1 - b$) enters only through the single impurity T matrix, which reduces in the 'pure ant' limit ($b = 1$) to $T(z) = 1/[4(1 - J(z))]$ and in the 'pure termite' limit ($b \to -\infty$) to $T(z) = 1/(4J(z))$.

The static diffusion coefficient is obtained by setting $z = 0$ in (8), resulting in the following density expansion:

$$D = \Phi(0) = \tfrac{1}{4}(1 + \alpha_1 c + \alpha_2 c^2 + O(c^3)). \tag{9}$$

As a consequence of self-duality the expansion coefficient satisfies $\alpha_1(1/\sigma) = -\alpha_1(\sigma)$ and $\alpha_2(1/\sigma) = \alpha_1^2(\sigma) - \alpha_2(\sigma)$.

The theory to linear order in the impurity concentration is identical to the EMA and yields $\alpha_1 = -2b/(2 - b) = -2(1 - \sigma)/(1 + \sigma)$. The coefficient of the $O(c^2)$ term can be expressed in rapidly converging lattice sums, containing the integrals $R_{\alpha\beta}(n, 0)$ that can be evaluated analytically for the relevant sites n close to the origin. The resulting value for the static diffusion coefficient $D(c, \sigma)$ is extrapolated to finite c values and plotted in figure 1 as a function of the concentration for several values of $\sigma = 1 - b \geqslant 0$. Using the symmetry (3) between high and low concentration we can also extrapolate from high concentrations, as shown in figure 1. The exact value $D(\tfrac{1}{2}, \sigma) = \sqrt{\sigma}D_0$ is also indicated. For the percolation case ($\sigma = 0$) the density expansion yields $D(c, 0) = (1 - 2c - 0.210\,75c^2 + \ldots)/4$ vanishing at $c \simeq 0.48$, whereas the exact percolation threshold for 2D bond percolation in a square lattice is $c_p = \tfrac{1}{2}$.

For σ values in the range $0.5 \leqslant \sigma < 1$ the $D(c, \sigma)$ values extrapolated from high and low impurity concentrations almost coincide and are therefore expected to give a good representation of $D(c, \sigma)$ over the whole range of impurity concentrations. The location of the exact value $D(\tfrac{1}{2}, \sigma) = \sqrt{\sigma}D_0$ for the half-filled lattice in figure 1 shows that for σ values, say, with $0.75 \leqslant \sigma \leqslant 0.9$ at least $O(c^3)$ terms are needed to have a reliable prediction for D in the range $0.4 \leqslant c \leqslant 0.6$. For the percolation case, where $\sigma = 0$, the ants are actually hopping on the percolation cluster far above threshold. If
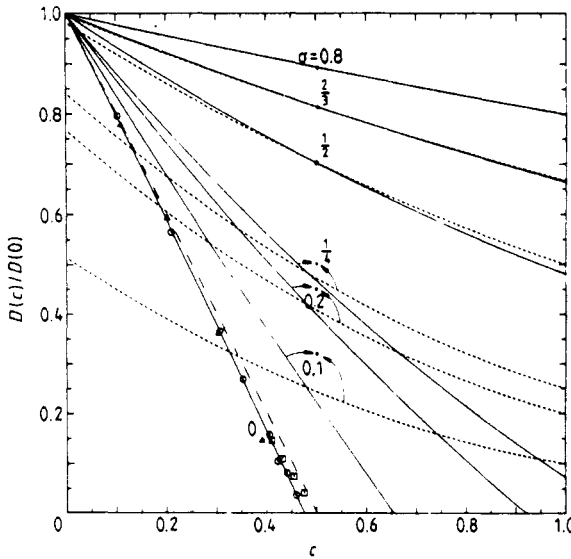

Figure 1. Diffusion coefficient $D(c)$ plotted against concentration c of impurity bonds for several values of the impurity conductance σ. The full curves are the extrapolations of the c expansion in (9); the broken curves are obtained from the symmetry relation $D(c, \sigma)D(1 - c, \sigma) = \sigma D^2(0)$ with $D(0) = \tfrac{1}{4}$ and extrapolation of the $(1 - c)$ expansion. The exact value $D(\tfrac{1}{2}, \sigma) = \sqrt{\sigma}D(0)$ is indicated by bold dots. For $\sigma = 0$ (bond percolation) the new results (full curve) are compared with effective medium theory ($- \cdot -$) and computer simulations (Kirkpatrick 1973).

the fraction of missing bonds increases, a fraction of the sites sit on islands surrounded by missing bonds. RW starting on these islands do not contribute to the static diffusion coefficient. As one needs to cut at least four bonds to make an island, this effect is of $O(c^4)$ in the concentration of missing bonds and does not yet interfere with our systematic density expansion. In the percolation case we have compared the results for the diffusion coefficient, obtained by Kirkpatrick from computer simulations with our results, exact to $O(c^2)$. Our $O(c)$ result for the percolation case, $D = \frac{1}{4}(1 - 2c)$, is identical to the effective medium approximation (EMA) which was already shown to be in reasonable agreement with the results from computer simulations and vanishes exactly at the percolation threshold, $c_p = \frac{1}{2}$, for bond percolation on a square lattice (Kirkpatrick 1973).

In order to study *the long time behaviour of the VACF* we need the dominant small-z singularity of $\Phi(z)$. We restrict ourselves first to $O(c)$ terms in (8), which is

$$\Phi(z) = \tfrac{1}{4}[1 - cb/(1 - bJ(z))] \tag{10}$$

representing the contributions from the 'repeated ring collisions'. The dominant small-z singularity of the ring collision integral (5) has the form

$$J(z) \simeq \tfrac{1}{2} \int_q q^2(4z + q^2)^{-1} \simeq \text{constant} + (1/2\pi)z \ln z \tag{11}$$

where we have used $\omega(q) \simeq \tfrac{1}{4}q^2(q \to 0)$. The singularity (11) induces a singularity in the VACF, given by

$$\Phi(z) \simeq \Phi(0) - (c/2\pi)b^2(2 - b)^{-2}z \ln z$$

which leads to a $1/t^2$ tail for long times. Identical results can be derived from the effective medium approximation (EMA), which is exact to $O(c)$ (see the review by Haus and Kehr (1986)). Using the EMA Odagaki *et al* (1983) have obtained the equivalent low frequency result for the AC conductivity, which becomes for a square lattice

$$\sum (\omega, \sigma) = \text{Re } \Phi(i\omega) \simeq \tfrac{1}{4}[1 + |\omega|c(1 - \sigma)^2/(1 + \sigma)^2]$$

with $\sigma = 1 - b$. We quote here the dominant long time behaviour for a d-dimensional cubic lattice, exact to $O(c)$, which can be obtained in a similar manner:

$$\phi(t) \simeq -\tfrac{1}{2}[c\pi b^2/(d - b)^2](d/2\pi t)^{1 + d/2} \qquad (t \to \infty). \tag{12}$$

Something peculiar happens as $b \uparrow 1$ in the one-dimensional case where the percolation threshold is at $c_p = 0$. For that case Denteneer and Ernst (1984) have calculated the exact long time tail for an arbitrary distribution of *non-vanishing* random variables ψ_n, defined below (2). In the present case the $\{\psi_n\}$ are distributed according to (with $b < 1$)

$$f(\psi) = c\delta(\psi - 1 - b) + (1 - c)\delta(\psi - 1).$$

The exact result for the long time tail in the VACF then becomes

$$\varphi(t) \simeq -\tfrac{1}{4}b^2c(1 - c)(1 - b + bc)^{-2}(D/\pi)^{1/2}t^{-3/2}$$

with $D^{-1} = 2[1 + bc/(1 - b)]$. This formula clearly shows that the density expansion breaks down if c and $1 - b$ are both small such that $1 - b \lesssim c$. However, for $c < 1 - b$ the above exact result reduces to (12). Next we consider the contribution to the long time tail of the VACF from scattering by two impurities. In order to extract from (8) the leading small-z singularity in $O(c^2)$ it is convenient to introduce the shorthand

notation $\delta A(z) \equiv A(z) - A(0)$ for the dominant small-z singularity which appears to be proportional to $z \ln z$ in the context of this paper. Using essentially the same arguments as in (11) one finds that the functions $R_{\alpha\beta}(n, z)$, appearing in (8), behave as

$$\delta R_{xx}(n, z) = [R_{xx}(n, 0) + 1][b/(2 - b)\pi]z \ln z$$

$$\delta R_{xy}(n, z) = R_{xy}(n, 0)[b/(2 - b)\pi]z \ln z.$$

Because of the slow convergence of the lattice sums, representing the small-z singularity in (8), it is required that part of these sums are calculated analytically before the remaining part can be evaluated numerically. The required integrals $R_{\alpha\beta}(n, 0)$ are the same as those occurring in α_2 of (9), and the final lattice sums converge again very rapidly. We thus obtain the long time tail $\phi(t) \simeq -\beta(c, \sigma)/t^2 (t \to \infty)$ with

$$\beta(c, \sigma) = (1/2\pi)b^2(2 - b)^{-2}c(1 + c\beta_1) + O(c^3). \tag{13}$$

The analytic expression for β_1, which will not be quoted here, exhibits the symmetry property $\beta(c, \sigma) = \beta(c, 1/\sigma)$. The reason is that $\beta(c, \sigma)$ depends on σ only through $|T(0)|$, defined in (5). Only in the two-dimensional case this quantity is invariant under the substitution $\sigma \to \sigma' = 1/\sigma$. The same mathematical property guarantees the symmetry relations of self-duality, discussed below (9), for the bond problem on the square lattice. Ernst (1987) relates this symmetry directly to self-duality. Of course, we also have the general symmetry (3), which implies $\beta(c, \sigma) = \beta(1 - c, 1/\sigma)$. In combining both symmetries we find the relation $\beta(c, \sigma) = \beta(1 - c, \sigma)$. We have extrapolated the functional form (13) of $\beta(c, \sigma)$ to finite c values and plotted it in figure 2 for high and low concentrations at several values of the conductivity ratio $\sigma = 1 - b$.
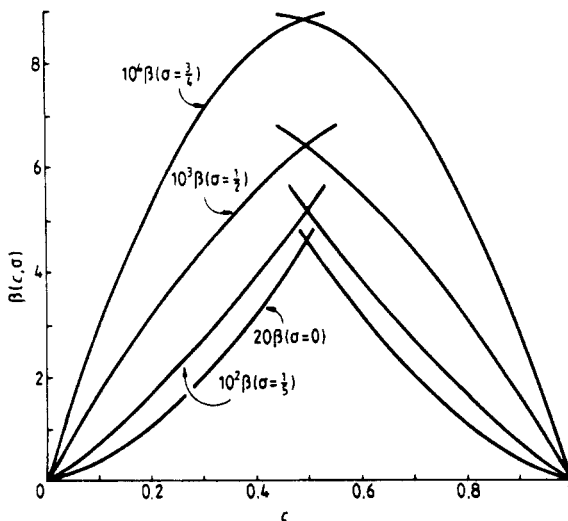

Figure 2. The coefficient $\beta(c, \sigma)$ of the long time tail of the VACF, $\varphi(t)$, plotted against impurity concentration c where $t^2\varphi(t) \simeq -\beta(c, \sigma)$ as $t \to \infty$. The curves for $c < 0.5$ are obtained from the low density series (13). The results at high densities are obtained from the relation $\beta(c, \sigma) = \beta(1 - c, \sigma)$, as implied by the symmetries of homogeneity and self-duality. There does not seem to be a meaningful interpolation between high and low densities as in figure 1.

The result for the long time tail in these lattice models, $\phi(t) \simeq -\beta/t^2$, is rather similar to the results for deterministic Lorentz models (Ernst *et al* 1984), where backscattering or cage effects cause *negative* velocity correlations with very long memory. The ant and termite models have the advantage that much more detailed analytic information can be obtained for these models than for the Lorentz gas, since the kinetic theory for the latter model is much more complicated. The blind ant on the square lattice with bond disorder and also with site disorder (Nieuwenhuizen *et al* 1986) are the only Lorentz-type models with dimensionality $d > 1$ for which the coefficient $\beta(c, \sigma)$ of the long time tail in the VACF could be calculated, exact to quadratic order in the density of scatterers. The plot in figure 2 also suggests that $\beta(c, \sigma)$ has a strong dependence on the impurity concentration. Since we do not know the exact value of $\beta(c, \sigma)$ for $c = \frac{1}{2}$, as was the case in figure 1, it is hard to tell how to interpolate between the high and low density results.

There is a further advantage over Lorentz gases: for our hopping model on a square lattice the ring collision integral $J(z)$ in (5) can be calculated explicitly in terms of the complete elliptic integral $K(x)$ of the first kind, in very much the same way as done by Morita and Horiguchi (1971) for 2D lattice Green functions with the result $J(z) = \frac{1}{2} - \pi^{-1} z (1+z)^{-1} K[(1+z)^{-2}]$. Its known asymptotic properties enable us to find subleading asymptotic corrections in (12), namely

$$\phi(t) \simeq -cb^2 [2\pi(2-b)^2 t^2]^{-1} \{1 + 4b[\pi(2-b)]^{-1} t^{-1} \ln(t/\tau_0) + O(t^{-2}(\ln t)^2)\} \tag{14}$$

where

$$\tau_0 = \tfrac{1}{8} \exp[\tfrac{3}{2} - \gamma + \pi/4 - \pi/(2b)] \tag{15}$$

with Euler's constant $\gamma \simeq 0.5772$. In fact, using the properties of $K(x)$ one can numerically invert the Laplace transform of (10) for *all times*. These $O(c)$ results for the VACF are plotted in figure 3 for $\sigma = 0$ and $\sigma = \frac{1}{2}$ and compared with the leading tail $\sim t^{-2}$ in (12) and the better approximation (14). The figure clearly shows that the VACF has not yet reached its pure asymptotic behaviour and that *one cannot see the pure t^{-2} tail in the time interval below 60 collision times*. This time interval covers the typical range where one has searched in vain for the tail $\phi(t) \sim -t^{-2}$ in computer simulations on Lorentz gases (Alder and Alley 1983). In this time interval we have tried to fit the exact time dependence, predicted by (14), to the function $\varphi(t) = -B/t^2$ or, equivalently, its time integral, defined in (1), to the function $\tilde{D}(t) = D + B/t$. This was done by plotting $\tilde{D}(t)$, obtained from (14), against $1/t$ and by measuring the apparent slope B. The result at $\sigma = 1$ and $\sigma = \frac{1}{2}$ was $B \simeq 1.9\beta$ and $B \simeq 1.5\beta$ respectively. This observation may explain why the reported values of β, measured in computer simulations on the 2D Lorentz gas (see, e.g., Alder and Alley 1983), are typically 50–100% too large. Thus we have to conclude that comparison of computer simulations and kinetic theory calculations for Lorentz-type models is only feasible for the presently accessible time intervals, if at least the first asymptotic correction term as in (14) and, preferably, the complete time dependence of $\phi(t)$ are known theoretically. This has been shown convincingly in recent computer simulations by Frenkel (1986) for the long time tail of the VACF of a blind ant on a square lattice with site disorder (Nieuwenhuizen *et al* 1986).

We complete the information on the VACF by considering its *short time behaviour*. This could be done by taking the high frequency limit ($z \to \infty$) in (10), which yields $\Phi(\infty) = \frac{1}{4}(1 - bc)$. Thus, the VACF for very short times has the form $\phi(t) \simeq \frac{1}{4}(1 - bc)\delta_+(t)$, where $\delta_+(t)$ is a Dirac delta function normalised as $\int_0^\infty dt\delta_+(t) = 1$. However, these
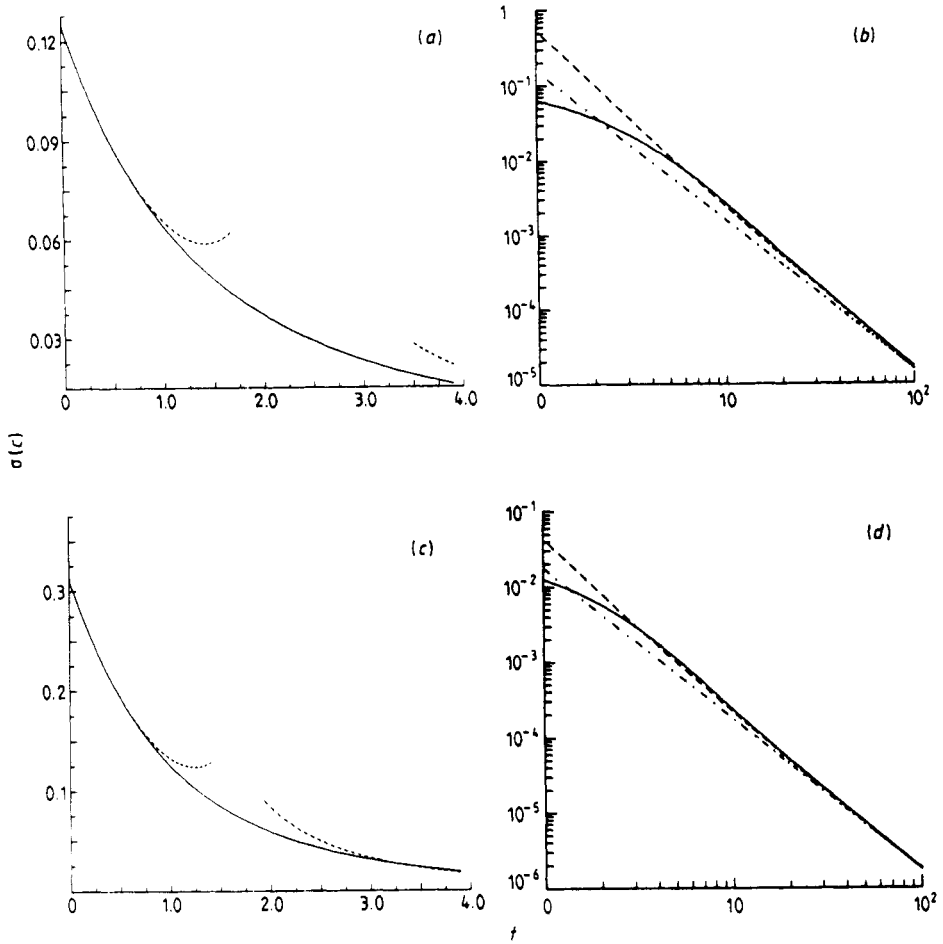
Figure 3. Velocity autocorrelation function $-\varphi_+(t)/c$ exact to $O(c)$ for all positive times $(t+0)$ at two values of the impurity conductance $\sigma = 1 - b$. In (a) and (c) the exact result (full curve) is compared with the short time expansion up to $O(t^4)$ and the long time expansion of (14) (broken curves). In (b) and (d) the exact values (full curve) of the VACF are compared with the long time tail $\varphi(t) \simeq -\beta(c)/t^2$ (— · —) and with $\varphi(t)$ in (14). $(a) B = 1, (b) B = 1, (c) B = 0.5, (d) B = 0.5$.

results are only consistent if $O(c^2)$ terms are neglected. *Exact results to all orders in c* can be obtained by calculating the response function (4) directly at short times:

$$\tilde{F}(q, t) = \langle \exp(tL) \rangle_{qq} = 1 - t\langle L_{qq} \rangle + \tfrac{1}{2}t^2 \langle L_{qq}^2 \rangle + \dots. \tag{16}$$

Here we will not write down the explicit expressions for these coefficients, but only discuss its consequences for the VACF:

$$\phi(t) \simeq D_E \delta_+(t) - (b^2/8)c(1-c) + (b^2/32)(5 - 2b - bc)c(1-c)t + O(t^2). \tag{17}$$

Here $D_E = \tfrac{1}{4}(1 - bc)$ is the short time limit of the time-dependent diffusion coefficient (1), $\tilde{D}(t) = \int_0^t d\tau \phi(\tau)$, which is referred to as the Enskog or mean-field value. It corresponds to a RW on a uniform lattice with a local 'collision frequency' or hopping rate $\tfrac{1}{4}(1 - bc)$, which is determined by the local 'free volume fraction' $(1 - bc)$. This is particularly obvious in the case of percolation (hard scatterers), where $b = 1$, so that

$(1-c)$ is the fraction of impurity-free bonds. There is no correlation between the velocities at two different times (no memory) of a RW on a uniform lattice, as shown by $D_E \delta_+(t)$.

However, the VACF of a RW on a random lattice exhibits for finite times the so-called 'cage effect', referring to the existence of memory effects leading to negative velocity correlations. Here $\phi_+(t) \equiv \phi(t) - D_E \delta_+(t)$ is, in fact, negative for all times. This bond model (together with the corresponding site version) is the only non-one-dimensional Lorentz-type or liquid-type model for which an exact prediction (to $O(c)$) of the cage effect has been given. The cage effect is the combined effect of individual hoppings on the lattice ('uncorrelated binary collisions' in kinetic theory language) and one or more returns to the same impurity ('repeated ring collisions'). The negative short time part $\phi_+(0) = -\tfrac{1}{8}c(1-c)b^2$ can be explained in a quantitative manner from the notion that a RW on a random lattice has an increased probability of being backscattered (Ernst 1986, Haus and Kehr 1986).

Finally it is instructive to consider the time-dependent diffusion coefficient $\tilde{D}(t)$ in (1). Its short time value $\tilde{D}(0+) = D_E$ was discussed above. Its long time value is determined by the cage effect, which substantially reduces the short time value as shown in figure 4, where the normalised time-dependent diffusion coefficient is defined as

$$d(t) = (\tilde{D}(t) - D_E)/(D_E - D_0)$$

$$= \int_0^t d\tau\, \phi_+(\tau)/(bcD_0) \tag{18}$$

with $D_0 = \tfrac{1}{4}$. If we write the relative difference, $\varepsilon = 1 - D(c)/D(0) = \varepsilon_E + \varepsilon_{RR} + O(c^2)$, then we have for the Enskog value $\varepsilon_E = bc$ and for the repeated ring (cage) contribution
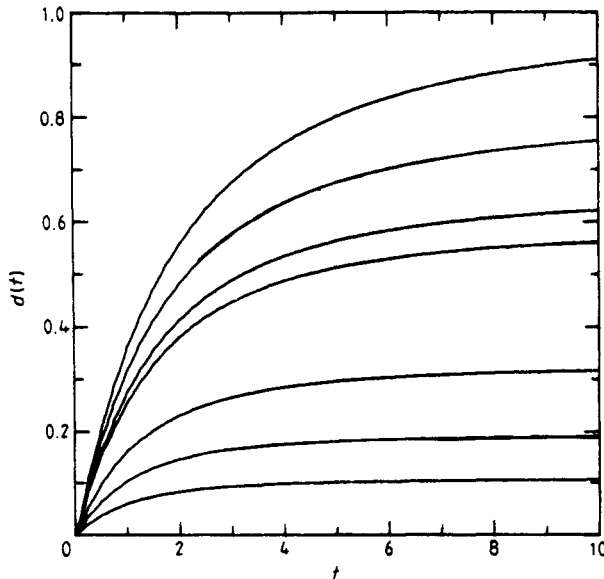

Figure 4. Decrease $d(t)$ in time of the time-dependent diffusion coefficient $\tilde{D}(t)$, defined in (18), for several values of the conductance σ of the impurity bonds (from top to bottom $\sigma = 0, \tfrac{1}{10}, \tfrac{1}{3}, \tfrac{1}{4}, \tfrac{1}{2}, \tfrac{2}{3}, \tfrac{4}{5}$).

$\varepsilon_{RR} = b^2 c/(2-b)$ with $\varepsilon_{RR}/\varepsilon_E = b/(2-b)$ with $b \leq 1$. For hard scatterers (percolation, $\sigma = 0$) and superconducting bonds ($\sigma \to \infty$) the cage effect is obviously the strongest. It also follows from $\phi(t) \simeq -\beta/t^2 (t \to \infty)$ that the diffusion coefficient $\tilde{D}(t) \simeq D + \beta/t(t \to \infty)$ approaches its static value only very slowly.

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