Journal of Statistical Physics, Vol. 48, Nos. 3/4, 1987

Time-Dependent Correlation Functions for Random Walks on Bond Disordered Cubic Lattices

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Received January 21, 1987

For hopping models on cubic lattices with a fraction c of impurity bonds, time-dependent transport properties and correlation functions (long-time tails) are calculated through a systematic c-expansion (in the percolation literature referred to as "high-density expansion"), using a method developed in an earlier paper. The time-dependent diffusion coefficient, velocity autocorrelation function (VACF), and Burnett functions are calculated exact to $O(c)$ for all t, and exact to $O(c^2)$ for long times only. A comparison is made with the results of the effective medium approximation, and numerical results are given for the square lattice.

KEY WORDS: Random walk on random lattice; bond percolation; random resistor network; Lorentz gas; velocity-correlation function.

1. INTRODUCTION

Kinetic theory methods have proven successful[1-3] for studying random walk (RW) problems on disordered lattices,[4] in which a fraction c of bonds or sites have been replaced at random by impurities. These methods produced systematic density expansions of transport properties and time correlation functions, exact to linear and quadratic order in the impurity concentration c for general dimensionality d.

For the special case of bond percolation (where an impurity corresponds to a blocked bond and where $p = 1 - c$ is the probability that a bond is open) the c-expansion yields what is referred to in the percolation literature as "high-density" expansion.[5] "Low-density" or p-expansions for the macroscopic conductivity and connectedness correlation functions are

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0022-4715/87/0800-0645$05.00/0

well developed[6] through a mapping of the percolation problem on the q-state Potts model with $q = 1$.[7] However, "high-density" or c-expansions only exist for the percolation probability and some moments of the cluster-size distribution function. For ac and dc transport properties *no exact* results are known except to linear order in $c = 1 - p$. Here one has to rely on approximations, such as the effective medium approximation (EMA).[8–10] For static transport coefficients in bond percolation problems away from the percolation threshold EMA happens to produce results in good agreement with existing computer simulations,[8] but EMA for frequency- or time-dependent phenomena has not been critically tested against exact results or computer simulations [see Ref. 3, below Eq. (6.92)].

In the present paper we study the so-called random barrier model (RBM)[9–13] or a random binary mixture of conductances σ and σ_0 on a hypercubic lattice, where the emphasis is on frequency and time dependence. Its static transport properties were already calculated in a previous paper,[3] referred to as paper I. A small selection of the time-dependent results for the square lattice has already been presented.[2]

Exact results on time dependence are of special interest not only for testing EMA, but also in view of the long-time tails $\sim t^{-1-d/2}$ in the velocity autocorrelation function (VACF) and similar tails in related time correlation functions, as occur in Lorentz gases and stationary random media.[14–18] In these models quantitative agreement is still lacking between computer simulations and theoretical predictions of time tails.[18]

A noteworthy exception is a recent simulation by Frenkel[19] on the VACF for a hopping model on a square lattice with a fraction of blocked sites.[20] Analytic results for that model[1] show that the pure long-time tail $\sim t^{-1-d/2}$ cannot be seen within time intervals accessible in modern computer simulations, say, up to 60–100 mean hopping times or mean free times. However, inclusion of the first subleading long-time behavior of the $O(c)$ correction [which is of relative $O(t^{-1} \ln t)$] shows quantitative agreement between analytic results and computer simulations for times from 20 to 60 mean hopping times.[19]

In the RBM to be considered in this paper, the analytic calculations are considerably simpler than those for the site-disordered model simulated by Frenkel. This makes it feasible to perform a numerical calculation of the VACF and other time correlation functions for *all times*, exact to $O(c^2)$ for 2D and 3D systems. In the site-disordered model, one has only been able to obtain the dominant long-time tail in $O(c^2)$.[1] The above properties make the present model a suitable candidate for computer simulations.[21]

Next we briefly recall that the RBM represents a RW model on a hypercubic lattice, in which a fraction c of the bonds chosen at random are

replaced by "impurity" bonds. The jump rates σ ("conductances") associated with an impurity bond may be larger or smaller than those of the host lattice bonds $\sigma_0 = 1$ (in appropriate units). The σ values range from $\sigma = 0$ (percolation, ants in a labyrinth problem)[22] via random resistor networks to termite problems where $\sigma \gg 1$.[4,11,22] The model can be described by the master equation

$$\dot{p}_n = \sum_{\rho} w_{n,n+\rho}(p_{n+\rho} - p_n) \tag{1.1}$$

Here $w_{n,n+\rho} = w_{n+\rho,n}$ is the transition rate to jump from site n to its nearest neighbor (n.n.) site $n+\rho$, defined as

$$w_{n,n+\rho} = (1/2d)(1 - bc_{n,n+\rho}) \tag{1.2}$$

The parameter σ (with $\sigma = 1 - b > 0$) represents the transition rate across a single bond, and $c_{n,n+\rho}$ is a random variable associated with the bond $(n, n+\rho)$ and defined as

$$c_{n,n+\rho} = \begin{cases} 1 & \text{with probability } c \\ 0 & \text{with probability } 1-c \end{cases} \tag{1.3}$$

The basic quantity is the response function:

$$F(q, z) = \int_0^\infty dt\, e^{-zt} \sum_{n,m} e^{iq(n-m)} \langle p(nt; m0) \rangle \tag{1.4}$$

where $\langle p(nt; m0) \rangle \equiv P_{n-m}(t)$ is the probability for a displacement $n-m$ of the RW, averaged over all impurity configurations $\{c_{n,n+\rho}\}$. In paper I this quantity has been systematically expanded in powers of c, and the coefficient of $O(c^m)$ is expressed in the form of cluster integrals (lattice sums) involving m impurities. The cluster integrals have the same structure as the m-tuple collision integrals in kinetic theory.[23] Derivatives of (1.4) with respect to q_x yield the moments of displacement. They determine the time-dependent diffusion coefficient $\tilde{D}(t)$ and the Burnett coefficient $\tilde{B}(t)$:

$$\tilde{D}(t) = \tfrac{1}{2}(\partial/\partial t)\langle n_x^2 \rangle = \int_0^t d\tau\, \phi(\tau) \tag{1.5a}$$

$$\tilde{B}(t) = \tfrac{1}{24}(\partial/\partial t)[\langle n_x^4 \rangle - 3\langle n_x^2 \rangle^2] \tag{1.5b}$$

Their long-time limits $\tilde{D}(\infty)$ and $\tilde{B}(\infty)$, if they exist, determine the static transport coefficients. The lattice analog of the VACF is

$$\phi(t) = \langle v_x(0)\, v_x(t) \rangle = \tfrac{1}{2}(\partial/\partial t)^2 \langle n_x^2 \rangle \tag{1.6}$$

In paper I we obtained a formal expression [see I, Eq. (4.3)] for its Laplace transform, $\phi(z) = \mathscr{L}_z(\phi(t))$, exact to $O(c^2)$.

In generalized hydrodynamics one usually defines a frequency-dependent diffusion coefficient and (modified) Burnett coefficient $\Phi(z)$ and $\Delta(z)$, respectively, through the small-q behavior of the response function,[17,18] namely

$$F(q, z) = [z + q^2\Phi(z) - q^4\Delta(z)]^{-1} \qquad (q \to 0) \tag{1.7}$$

The quantity $\Phi(z)$ is in fact the Laplace transform of the VACF, and $\Phi(0) = \tilde{D}(\infty) \equiv D$ is the static diffusion coefficient. The ac conductivity is essentially given through $\Sigma(\omega) = \mathrm{Re}\,\Phi(i\omega)$. The ordinary and modified Burnett coefficients are also interrelated, as shown in Appendix A. Since we shall be discussing lattices with discrete translational and rotational symmetries, D and Φ are second-rank tensors, and B and Δ are fourth-rank tensors. A formulation appropriate for cubic lattices is given in Appendix A.

There exists an extensive literature on long-time tails in fluids[24] and in stationary random media, such as Lorentz gases and RW models on disordered lattices.[10,18,25] In fluids, the VACF has a positive tail $\sim t^{-d/2}$; in diffusive systems the tail is $\phi(t) \sim -\langle(\delta D)^2\rangle\, t^{-1-d/2}$, where mode coupling theories show that the coefficient is proportional to the variance in the spatially fluctuating diffusion coefficient.[18]

Long memory effects cause the Burnett coefficients to diverge in some cases.[18,26] In fluids $\tilde{B}(t) \sim t \ln t$ for $d = 2$ and $\tilde{B}(t) \sim \sqrt{t}$ for $d = 3$, whereas in diffusive systems $\tilde{B}(t) \sim \log t$ for $d = 2$ and $\tilde{B}(t)$ is finite for $d = 3$. The coefficients $\Delta(z)$ show similar singular behavior for $z \to 0$.

In paper I it was also shown that replacement of σ-bonds by σ_0-bonds and vice versa and a simultaneous rescaling of the time gives the symmetry relation $P_n(c, \sigma, t) = P_n(1 - c, 1/\sigma, \sigma t)$, implying

$$\begin{aligned} \tilde{D}(t, c, \sigma) &= \sigma\tilde{D}(\sigma t, 1 - c, 1/\sigma) \\ \phi(t, c, \sigma) &= \sigma^2\phi(\sigma t, 1 - \sigma, 1/\sigma) \end{aligned} \tag{1.8}$$

The formal expression for the VACF from paper I, exact to $O(c^2)$, is given by

$$\begin{aligned} \Phi(z) = D_0 - cT - c^2T\Big\{ & -2dJT + 4d^2T^2(J - J^2 + zJ') \\ & + \sum_{n \neq 0} R_{xx}^3(n, z)/[1 - R_{xx}(n, z)] \\ & + (d-1)\sum_{n=0} R_{xy}^4(n, z)/[1 - R_{xy}^2(n, z)]\Big\} \end{aligned} \tag{1.9}$$

where $D_0 = 1/2d$ and $J'(z) = dJ(z)/dz$ and

$$T(z) = (b/2d)/[1 - bJ(z)] \tag{1.10a}$$

$$J(z) = (1/d) \int_q \omega(q)/[z + \omega(q)] \tag{1.10b}$$

$$R_{\alpha\beta}(n, z) = T(z)\, G_{\alpha\beta}(n, z) \tag{1.10c}$$

$$G_{\alpha\beta}(n, z) = \int_q e^{-inq}(1 - e^{-iq_\alpha})(1 - e^{iq_\beta})/[z + \omega(q)] \tag{1.10d}$$

The average $\int_q$ is taken over the first Brillouin zone of a hypercubic lattice with $q = \{q_1 q_2 \cdots q_d\}$, namely

$$\int_q f(q) = (2\pi)^{-d} \int_{-\pi}^{\pi} dq_1 \int_{-\pi}^{\pi} dq_2 \cdots \int_{-\pi}^{\pi} dq_d\, f(q) \tag{1.11}$$

and

$$\omega(q) = d^{-1} \sum_{\alpha=1}^{d} (1 - \cos q_\alpha) \tag{1.12}$$

The paper is organized as follows: In Section 2 the short- and intermediate-time behavior of the VACF and time-dependent diffusion coefficient are calculated. Their long-time behavior is discussed in Section 3. Burnett coefficients are treated in Section 4 and Appendix A. The results obtained and their relevance for computer simulations and testing approximate theories are discussed in Section 5.

2. SHORT- AND INTERMEDIATE-TIME BEHAVIOR OF VACF

The VACF to linear order in the impurity concentration c is determined by summing all repeated visits of the RW to a single impurity site. This quantity in turn is determined by the probability of return to the origin on a uniform lattice, which is essentially the ring collision integral in kinetic theory.[16,17]

The Laplace transform of the VACF is explicitly given by [see (1.7) and (1.8)]

$$\Phi(z) = D_0\{1 - bc/[1 - bJ(z)]\} \tag{2.1}$$

It is convenient to write the ring collision integral (1.8) as

$$J(z) = d^{-1}[1 - zL(z)] \tag{2.2}$$

with

$$L(z) \equiv \int_q [z + \omega(q)]^{-1} = -L(-2-z) \tag{2.3}$$

and one easily verifies the above symmetry.

At short times we determine $\phi(t)$ from an expansion of (2.1) in powers of $1/z$ and a subsequent term-by-term Laplace inversion: $n!\, z^{-n-1}$ corresponds to t^n and 1 to $\delta_+(t)$, which is a Dirac delta function normalized as $\int_0^\infty dt\, \delta_+(t) = 1$. Since the integrals occurring in the z^{-1} expansion of (2.2) are elementary, we simply state the first few terms in the short-time expansion of the VACF:

$$\begin{aligned}\phi(t) = {} & D_0(1-bc)\,\delta_+(t) - D_0 cb^2/d + tD_0 c(b^2/2d^2)(2d+1-2b) \\ & - t^2 D_0 c(b^2/4d^3)(2d^2+3d-4db-2b+2b^2) + O(t^3)\end{aligned} \tag{2.4}$$

If so desired, one can easily extend it to higher orders in t. We only quote this expression because it has been used in testing our numerical procedure for calculating the inverse Laplace transform of $\Phi(z)$ and the results are shown as dashed curves in all figures at short times. In Eq. (6.10) of paper I we have, in fact, already obtained the exact short-time behavior of $\phi(t)$ at arbitrary impurity concentration up to $O(t)$ terms included for the RBM on a d-dimensional lattice. For the random chain ($d=1$) the short-time expansion at *arbitrary impurity concentration* is actually known up to $O(t^3)$ term included [see Eq. (16) of Ref. 27] if one calculates the moments of the random variable (1.2) using (1.3), namely

$$\langle (w - \langle w \rangle)^n \rangle = (b/2)^n [c(c-1)^n + (1-c)\, c^n] \tag{2.5}$$

In order to determine the VACF at finite times we have to invert the Laplace transform of (2.1). We first observe that $\Phi(z) \approx \Phi(\infty) + O(z^{-1})$ as $z \to \infty$, since $J(z) \approx z^{-1} + O(z^{-2})$. Hence, the inverse Laplace transform of the remainder $\Phi(z) - \Phi(\infty)$ exists, and we define

$$\begin{aligned} D_E &\equiv \Phi(\infty) = D_0(1-bc) \\ \Phi_+(z) &\equiv \Phi(z) - \Phi(\infty) = -D_0 cb^2 J(z)/[1 - bJ(z)] \end{aligned} \tag{2.6}$$

Here D_E is the so-called Enskog value of the diffusion coefficient (also referred to as "short-time," "bare," or "high-frequency" value). The factor $(1-bc)$ in (2.6) is the effective free volume fraction or porosity of the lattice.

The VACF is now given by

$$\phi(t) = D_E \delta_+(t) + \phi_+(t)$$
$$\phi_+(t) = (2\pi i)^{-1} \int_{-i\infty+0}^{i\infty+0} dz\, \Phi_+(z) \exp(zt) \tag{2.7}$$

The contour along the imaginary axis can be closed in the left half-plane ($\mathrm{Re}\, z < 0$) by a large semicircle. The contributions come from a possible pole $\phi_p(t)$ and from a branch cut $\phi_b(t)$ extending along the negative real axis with $\mathrm{Re}\, z \in [-2, 0]$. The branch cut is determined by the interval where the denominator $z + \omega(q)$ in the integral (2.3) is vanishing. The pole of $\Phi_+(z)$ is determined by the zero of the denominator in (2.6):

$$bJ(z) = 1 \tag{2.8}$$

where $b \leqslant 1$, except for the one-dimensional case, where $b < 1$.

The analysis is best illustrated by first considering the 1D case. There, calculation of the ring collision integral (2.2) yields

$$J(z) = 1 - [z/(2+z)]^{1/2} \tag{2.9}$$

so that

$$\Phi_+(z) = \tfrac{1}{2}cb\{1 - [z/(2+z)]^{1/2}\}/\{1 - b^{-1} - [z/(2+z)]^{1/2}\} \tag{2.10}$$

As far as poles are concerned, Eq. (2.8) only has a solution for $b < 0$, corresponding to impurities with a transition rate $\sigma = 1 - b$ that is larger than the transition rate $\sigma_0 = 1$ of the host lattice. The pole is located on the negative real axis at $z_p = -2(1-b)^2/(1-2b)$ to the left of the branch cut, since $z_p < -2$ for $b < 0$. The resulting pole contribution to the VACF is

$$\phi_p(t) = c[D_0/J'(z_p)] \exp(z_p t) \tag{2.11a}$$
$$= c[2(1-b)b^3/(1-2b)^2] \exp(z_p t) \tag{2.11b}$$

where the prime on $J(z)$ denotes a derivative.

The contribution from the branch cut is

$$\phi_b(t) = \pi^{-1} \int_0^2 dx\, e^{-xt}\, \mathrm{Im}\, \Phi_+(-x - i0) \tag{2.12a}$$
$$= (c/\pi)\, e^{-t} \int_{-1}^{1} dx\, e^{-xt} (1-x^2)^{1/2}/[\alpha^2 + 1 + (\alpha^2 - 1)x] \tag{2.12b}$$

with $\alpha = 1 - b^{-1}$. The second expression follows from (2.10) and (2.12a)

after shifting the integration variable and symmetrizing the integrand. The VACF is then given by

$$\phi_+(t) = \begin{cases} \phi_b(t), & b > 0 \\ \phi_p(t) + \phi_b(t), & b < 0 \end{cases} \tag{2.13}$$

As a test one might calculate (2.12b) at $t = 0$. After the substitution $y = [(1+x)/(1-x)]^{1/2}$ the integral can be evaluated by contour integration and yields

$$\phi_b(0) = \begin{cases} \frac{1}{2}cb^2, & b > 0 \\ \frac{1}{2}cb^2/(1-2b)^2, & b < 0 \end{cases} \tag{2.14}$$

One verifies with the help of (2.11) and (2.14) that $\phi_+(0) = cb^2/2$ for all $b < 1$ in accordance with (2.4) for $d = 1$.

Next, we consider the general case where the ring collision integral is determined by (2.2). The pole in $\Phi_+(z)$ is determined by solving (2.8). It is located on the negative real axis outside the branch cut. Thus, we set $z = -2 - \zeta$ ($\zeta > 0$) and rewrite (2.8) using $L(z) = -L(-z-2)$, with the result

$$L(\zeta) = (1 - d/b)/(2 + \zeta) \tag{2.15}$$

We use a graphical solution method: observe that $L(\zeta)$ is positive and monotonically decreasing for $\zeta \geqslant 0$. Furthermore, $L(0)$ is divergent for $d = 1$ and 2, and $L(0) < \infty$ for $d \geqslant 3$, and decays like $L(\zeta) \approx \zeta^{-1} - \zeta^{-2} + \cdots$ as $\zeta \to \infty$. For Eq. (2.15) to have a solution, its rhs must also be positive. Consequently, $b < 0$ [see (1.2)]. As $b \uparrow 0$, the rhs grows beyond all bounds, and there is only a solution for $d = 1, 2$. For $d \geqslant 3$ there exists only a solution if $b \leqslant b_0(d) = d/[2L(0) - 1]$ where $L(0)$ for $d = 3$ is a Watson integral, given by $L(0) \approx 1.516386$.[30]

As $b \to -\infty$, the root $\zeta_p \to \infty$ and can be determined analytically as $\zeta_p \simeq -b/d + O(b^0)$ or $z_p = -2 - \zeta_p = b/d + O(b^0)$ as $b \to -\infty$. The residue in (2.11a) can be calculated straightforwardly, yielding

$$\phi_p(t) \approx -c(d^2/2b^2)\exp(tb/d), \qquad b \to -\infty \tag{2.16}$$

As $b \uparrow 0$, the root $\zeta_p \downarrow 0$ for one and two dimensions. The 1D case was already solved exactly in (2.11b). For the 2D case, where $L(z) \approx -\pi^{-1}\ln(z/8)$ as $z \to 0$, one finds $\zeta_p \approx 8\exp(2\pi/b)$ and the pole contribution to the VACF is

$$\phi_p(t) \approx -c(\pi\zeta_p/4)\exp[-(2 + \zeta_p)t], \qquad b \to -\infty \tag{2.17}$$

For finite, negative b values, Eq. (2.8) has to be solved numerically. From now on we restrict ourselves to the two-dimensional case, where the ring integral can be calculated analytically in terms of $L(z)$ [see I, (C5)]:

$$L(z) = (2/\pi) \int_0^{\pi/2} dt\, [(1+z)^2 - \cos^2 t]^{1/2}$$

$$= (2/\pi)(1+z)^{-1} K[(1+z)^{-2}] \tag{2.18}$$

Here $K(x)$ is the complete elliptic integral of the first kind [see Eq. (17.3.1) of Ref. 31]. The residue in (2.11a) is obtained from the relation

$$L'(z) = -[z(2+z)]^{-1}(2/\pi)\, E[(1+z)^{-2}] \tag{2.19}$$

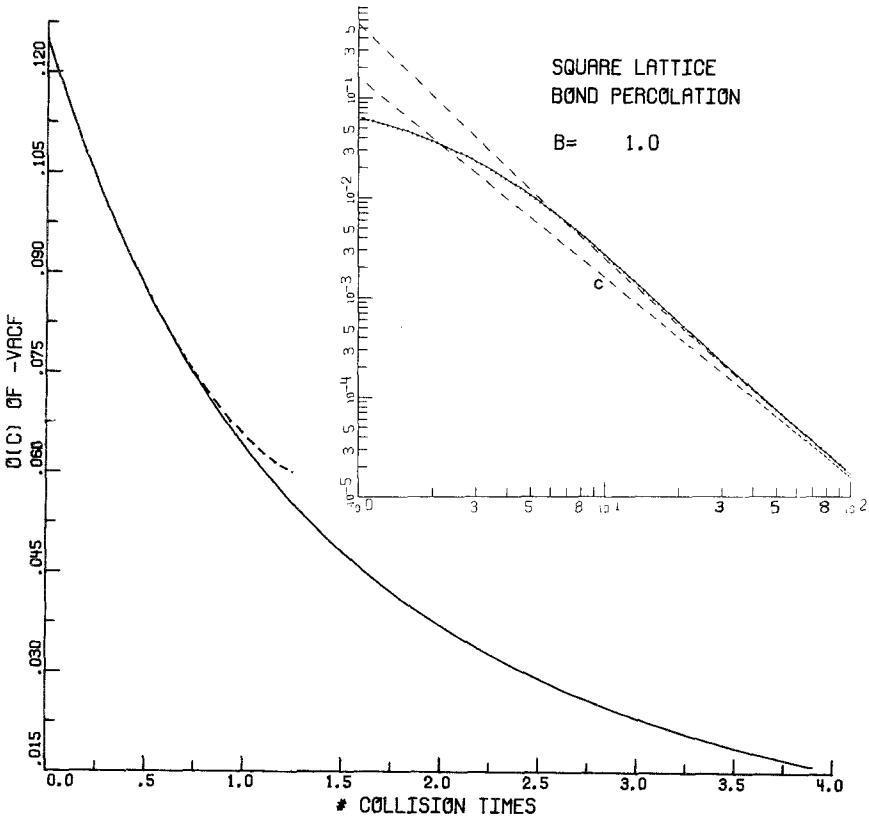

Fig. 1. Plot of $-$VACF $\phi_+(t)$ in (2.7) to $O(c)$ as a function of the number of mean collision or hopping times t for parameter value $\sigma = 1 - b = 0$ (percolation, ants). The insert is a blowup of the long-time behavior. Dashed lines on the rhs represent the short-time approximation (2.4); dashed lines on the lhs represent the pure long-time tail (3.4) (straight line, labeled c) or inclusion of the subleading term (3.7).

where $E(x)$ is the complete elliptic integral of the second kind [see Eq. (17.3.3) of Ref. 31]. The above equation can be derived from Eqs. (17.3.9, 10), (15.3.3), and (15.2.15) of Ref. 31. The pole contribution for $d=2$ becomes then

$$\phi_p(t) = -(c\pi/4)\, e^{z_p t} \times \{(z_p+1)^{-1} K[(z_p+1)^{-2}] - (z_p+2)^{-1} E[(z_p+1)^{-2}]\}^{-1} \tag{2.20}$$

for $b<0$, whereas $\phi_p(t)=0$ for $0<b\leqslant 1$.

To determine the contribution from the branch cut (2.12a), we split (2.18) for $z=-x+i0$ into its real and imaginary parts. Let $t_0=$

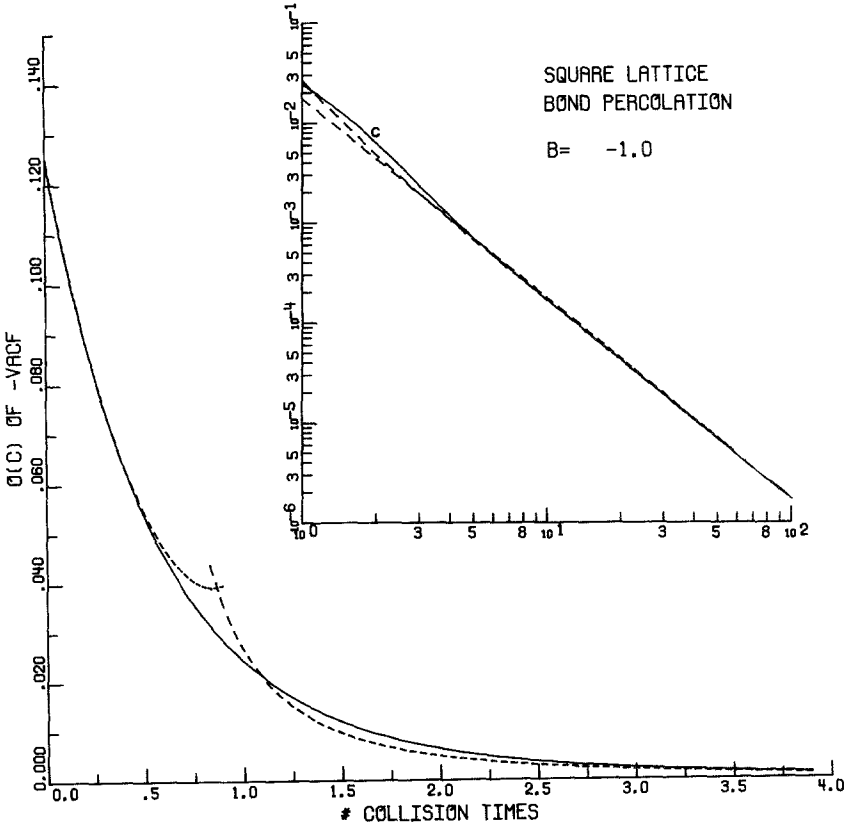

Fig. 2. Same as Fig. 1, for $\sigma=1-b=2$.

arcos$(1-x)$ be the angle where the argument of the square root in (2.18) changes sign. Then it follows for $0 \leqslant x \leqslant 1$

$$L(-x \pm i0) = L_1(x) \mp iL_2(x)$$
$$L_1(x) = (2/\pi)\, K(\cos^2 t_0) = (2/\pi)\, K[(1-x)^2]$$
$$L_2(x) = (2/\pi)\, K(\sin^2 t_0) = (2/\pi)\, K[x(2-x)]$$

For $x \in [1, 2]$ one can derive analogous relations using the symmetry $L(z) = -L(-z-2)$. We omit the detailed analysis and only quote the result:

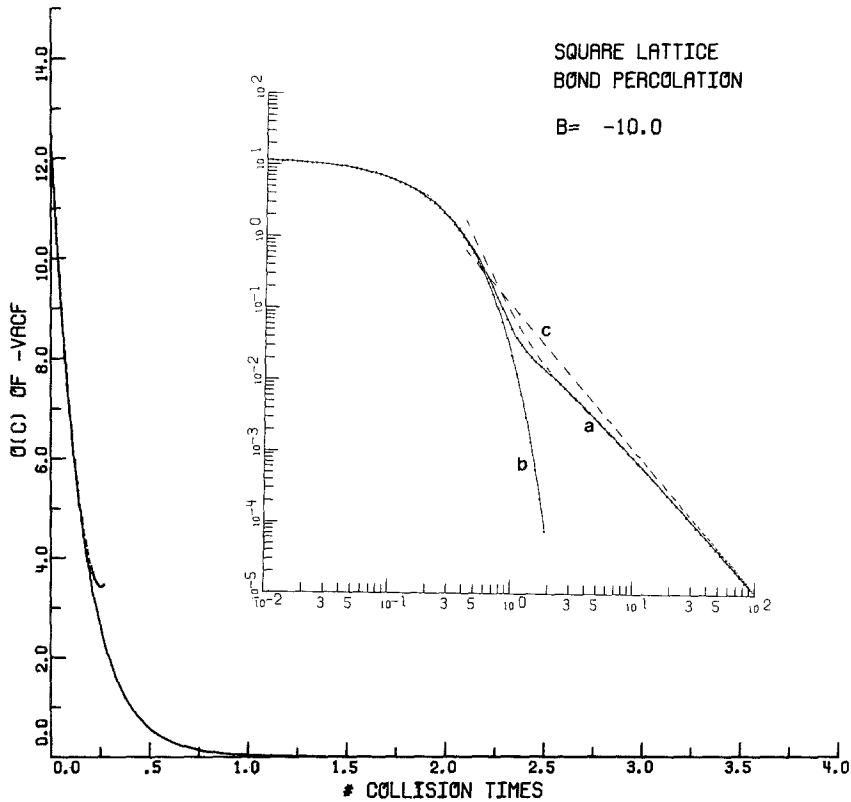

Fig. 3. Same as Fig. 1, for $\sigma = 1 - b = 11$ (superconducting bonds, termites). The time scale $(0.01 < t < 100)$ differs from that in Figs. 1 and 2. The curves labeled a and b represent, respectively, $\phi_+(t)$ and $\phi_p(t)$, defined in (2.13) and discussed in Section 5.

$$\phi_b(t) = (cb^2/2\pi) \int_0^1 dx\, xL_2(x)\, e^{-xt}$$
$$\times \{[2-b-bxL_1(x)]^2 + [bxL_2(x)]^2\}^{-1}$$
$$+ (cb^2/2\pi) \int_0^1 dx\, (2-x)\, L_2(x)\, e^{(x-2)t}$$
$$\times \{[2-b-b(2-x)\, L_1(x)]^2 + [b(2-x)\, L_2(x)]^2\}^{-1} \quad (2.21)$$

The final result for the VACF to $O(c)$ in the 2D case therefore equals $\phi_b(t)$ for $b>0$, and for $b<0$ one has $\phi_+(t) = \phi_p(t) + \phi_b(t)$. The expressions in (2.20) and (2.21) have been evaluated numerically and the results are shown in Figs. 1–3 for the values of $b=1$, -1, -10. The plots show $-\phi_+(t)$ (solid line) defined in (2.7) and denoted by $O(c)$ of $-\mathrm{VACF}(t)$ on the vertical axis of the graphs. The dashed lines on the left represent the short-time behavior obtained from (2.4) with up to $O(t^4)$ terms included.

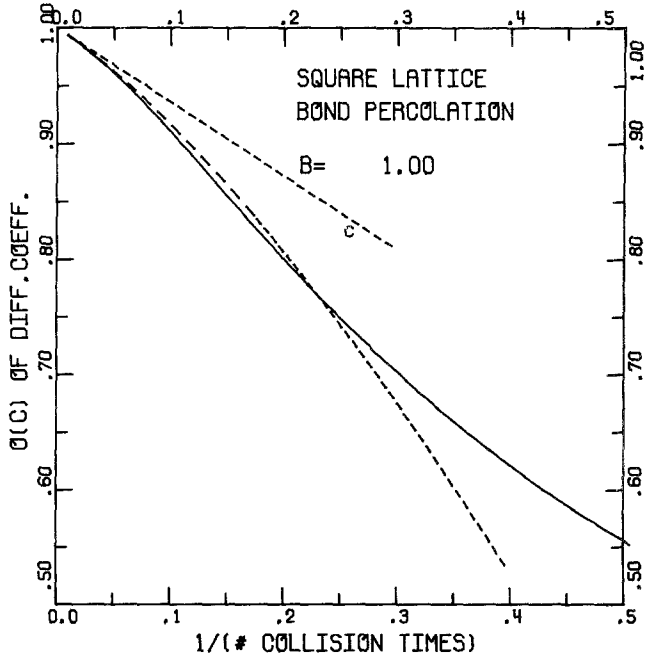

Fig. 4. Reduced time-dependent diffusion coefficient $d(t)$, defined in (2.22), to $O(c)$ as a function of $1/t$, the inverse number of collision or hopping times at parameter value $\sigma = 1-b=0$. The curved dashed line represents the long-time approximation (3.8) and the straight dashed line represents the pure asymptotic tail $\tilde{D}(t) \simeq D + \beta/t$.

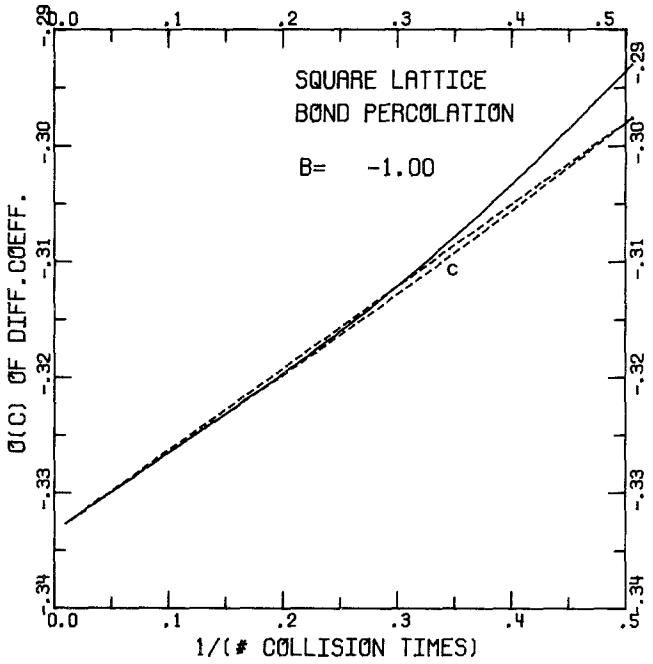

Fig. 5. Same as Fig. 4 at parameter value $\sigma = 1 - b = 2$.

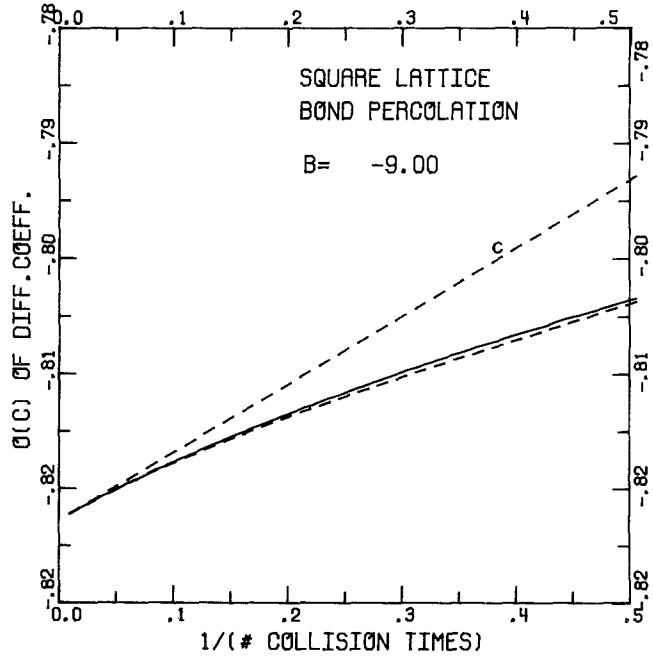

Fig. 6. Same as Fig. 4 at parameter value $\sigma = 1 - b = 10$.

The dashed lines on the right represent long-time behavior, to be discussed in the next section.

The time-dependent diffusion coefficient $\tilde{D}(t)$ defined in (1.5) is the inverse Laplace transform of $z^{-1}\Phi(z)$. It can be calculated along the same lines as $\phi(t)$, and the results are shown in Figs. 4–6. What is actually shown is the density-*independent* quantity

$$d(t) = [D(t) - D_E]/(D_E - D_0) = (bcD_0)^{-1} \int_0^t d\tau\, \phi_+(\tau) \qquad (2.22)$$

where $\tilde{D}(0+) = D_E = D_0(1-bc)$ with $D_0 = 1/4$ is the high-frequency or Enskog diffusion coefficient (2.6). The dashed lines represent short- and long-time approximations.

3. LEADING AND SUBLEADING LONG-TIME TAILS

3.1. $O(c)$ Contributions

In view of the long-standing interest and difficulties[14–18] around long-time tails in Lorentz-gas-type models, we devote a separate section to asymptotic long-time behavior of the VACF. In the first subsection we consider the contributions linear in the impurity concentration, and in the next subsection the $O(c^2)$ contributions.

Long-time tails of the VACF are determined by the singularities in $\Phi(z)$ that are closest to the origin. In the $O(c)$ terms of (2.1) the dominant singularity is a branch point at the origin, as discussed in Section 2. Let $\delta A(z)$ denote the singular part of $A(z)$ as $z \to 0$. Then we determine $\delta J(z)$ for the ring collision integral $J(z)$. This singularity is determined by the small-q integration region in (2.3), where

$$\omega(q) \simeq D_0 \sum_{\alpha=1}^{d} (q_\alpha^2 - q_\alpha^4/12 + \cdots)$$

In time language, the long-time tail or singular part follows from (2.2) and (2.3) as

$$\begin{aligned} \delta\tilde{J}(t) &\simeq -d^{-1}(2\pi)^{-d}(d/dt) \int d^{(d)}q \exp\left(-tD_0 q^2 + tD_0 \sum_\alpha q_\alpha^4/12 + \cdots\right) \\ &\simeq \tfrac{1}{2}(d/2\pi)^{d/2}\, t^{-1-d/2}[1 + d(d+2)/8t + \cdots] \end{aligned} \qquad (3.1)$$

or in Laplace language as

$$\delta J(z) \simeq \tfrac{1}{2}(d/2\pi)^{d/2}\,\Gamma(-d/2)\,z^{d/2}(1 - zd/4 + \cdots) \tag{3.2}$$

with $z \to 0$.

We have used the Tauberian theorems, listed in Table I, relating the large-t behavior of a function $\tilde{f}(t)$ to the small-z behavior of its Laplace transform $f(z)$. In fact, (3.1b) applies to the odd d values; for even d values, Eq. (3.1b) is defined through a limiting operation with $d = 2n + \varepsilon$ as $\varepsilon \to 0$ and $n =$ integer. The singularity $\delta J(z)$ induces a singularity in $\Phi(z)$ to dominant order given by

$$\delta\Phi(z) \approx -\tfrac{1}{2}cdb^2(d-b)^{-2}\,\delta J(z), \qquad z \to 0 \tag{3.3}$$

Thus, the dominant long-time tail of the VACF to $O(c)$ is

$$\phi(t) \approx -\tfrac{1}{2}[c\pi b^2/(d-b)^2](d/2\pi t)^{1+d/2}, \qquad t \to \infty \tag{3.4}$$

To have an estimate of the time scale on which the tail (3.4) is dominant, we determine the first correction to (3.4). In fact, for the 1D case, the leading and subleading long-time tail of the VACF is known exactly for arbitrary impurity concentration[27–29] and has the form

$$\phi(t) \simeq -\beta t^{-3/2}(1 + \tau_0/t + \cdots), \qquad t \to \infty$$

with

$$\beta = (4\sqrt{\pi})^{-1}\, b^2(1-b)^{1/2}(1-b+bc)^{-5/2}\, c(1-c)$$

$$\tau_0 = -3\pi a_1/\mu_2$$

where a_1 can be expressed in terms of moments μ_2, μ_3 and μ_4 (see Table 1, Ref. 28), defined as

$$\mu_n = D^n\langle(w^{-1} - \langle w^{-1}\rangle)^n\rangle = [c(1-c)^n + (1-c)c^n]\, b^n/(1-b+bc)^n$$

Table I. Tauberian Theorems

Small-z behavior	Large-t behavior
$\Gamma(-\alpha)z^\alpha$ $(\alpha \neq 0, 1, 2, \ldots)$	$t^{-\alpha-1}$
$(-)^{n+1} z^n \ln z$ $(n = 0, 1, 2, \ldots)$	$n!\, t^{-n-1}$
$-z(\ln z)^2$	$(2 \ln t + 2\gamma - 2)\, t^{-2}$
$z^2(\ln z)^2$	$(4 \ln t + 4\gamma - 6)\, t^{-3}$

Euler's constant $\gamma \simeq 0.5772$

Here $D = 1/\langle w^{-1} \rangle = (1-b)/(1-b+bc)$ is the exact diffusion coefficient for the 1D case. The second equality follows from Eq. (1.3).

In the 2D case we analyze the $O(c)$ term (2.1) in the VACF. The singular part of $J(z)$ is given by (3.2) with the appropriate limiting operation ($d = 2+\varepsilon$, $\varepsilon \to 0$). However, we also need the regular part of $J(z)$, correct to relative $O(z)$ [see I, Eq. (C7)]

$$J(z) \simeq 1/2 + (1/2\pi)(1 - z/2) z \ln(z/8) + \cdots \tag{3.5}$$

Combining (3.5) with (2.1) yields for the singular small-z behavior in the 2D case

$$\delta\Phi(z) \simeq -c(b^2/2\pi)(2-b)^{-2}\, z \ln(z/8)[1 - \tfrac{1}{2}z + (b/\pi)(2-b)^{-1}\, z \ln(z/8)] \tag{3.6}$$

and the corresponding long-time tails, exact to $O(c)$, can be read off from Table I:

$$\phi(t) \simeq -\beta t^{-2}[1 + (\tau_0/t)\ln(t/\tau_1) + \cdots] \tag{3.7a}$$

with

$$\begin{aligned} \beta &= c(b^2/2\pi)(2-b)^{-2} \\ \tau_0 &= (4b/\pi)(2-b)^{-1} \\ \tau_1 &= \tfrac{1}{8}\exp(3/2 - \gamma + \pi/4 - \pi/2b) \end{aligned} \tag{3.7b}$$

where $\gamma = 0.5772$ is Euler's constant. Note that the approach to the t^{-2} tail is of relative $O(t^{-1} \ln t)$, which is extremely slow. In Figs. 1–3 the long-time behavior and the asymptotic corrections (3.7) are compared with the intermediate-time behavior obtained numerically in Section 2.

The time dependence of the diffusion coefficient $\tilde{D}(t)$ defined in (1.5) is given by the inverse Laplace transform of $z^{-1}\Phi(z)$. Its asymptotic long-time behavior thus follows immediately from $z^{-1}\delta\Phi(z)$ and Table I. We only quote the result for the 2D case:

$$\tilde{D}(t) = D + (\beta/t)[1 + (\tau_0/2t)\ln(t\sqrt{e}/\tau_1) + \cdots] \tag{3.8}$$

where $D = \tilde{D}(\infty) = \frac{1}{4}[1 - 2bc/(2-b)]$ is the static diffusion coefficient to $O(c)$ [see I, (4.4)]. For dimensionality $d \geqslant 3$ the first correction term to the dominant singularity in the VACF is always of relative $O(z)$ and is contained in

$$\delta\Phi(z) \simeq -(cd/2)\, b^2[d - b + zbL(0)]^{-2}\, \delta J(z)$$

with $L(z)$ defined in (2.3). The long-time tail of VACF, exact to $O(c)$, and including its first correction follows then from Table I and (3.4) as

$$\phi(t) \simeq -\beta t^{-1-d/2}(1+\tau^{(d)}/t+\cdots) \tag{3.9a}$$

with

$$\begin{aligned} \beta &= (cd/4)\, b^2(d-b)^{-2}(d/2\pi)^{d/2} \\ \tau^{(d)} &= d(d+2)/8 + (d+2)\, bL(0)/(d-b) \end{aligned} \tag{3.9b}$$

3.2. $O(c^2)$ Contribution

Next, we consider the $O(c^2)$ contribution to the VACF in (1.7), restricting ourselves to the singular part. We investigate the singular part of the quantities appearing in (1.9). Starting with (1.10a), one finds

$$\delta T(z) \simeq 2dT^2(0)\, \delta J(z)$$

with δJ given in (3.1). The singular part of $R_{\alpha\beta}(n, z) = T(z)\, G_{\alpha\beta}(n, z)$ can be determined using similar arguments as in (3.1a) and one obtains

$$\delta\tilde{G}_{\alpha\beta}(n, t) = \delta_{\alpha\beta}\delta\tilde{G}_{xx}(0, t) \simeq 2d\delta_{\alpha\beta}\delta\tilde{J}(t)$$

This implies for $R_{\alpha\beta}(n, z)$ in (1.10c)

$$\begin{aligned} \delta R_{xx}(n, z) &\simeq 2dT(0)[R_{xx}(n, 0)+1]\, \delta J(z) \\ \delta R_{xy}(n, z) &\simeq 2dT(0)\, R_{xy}(n, 0)\, \delta J(z) \end{aligned}$$

For the dominant small-z singularity of the $O(c^2)$ term in (1.9) one finds after a lengthy calculation using the results of Appendix B

$$\delta\Phi(z) \simeq -(c/2)\, db^2(d-b)^{-2}(1+\beta_1 c+\cdots)\, \delta J(z) \tag{3.10}$$

where the coefficient is given by

$$\begin{aligned} \beta_1 = {} & \tfrac{1}{2}d(d-b)^{-2}[b^2(4-d)+b(d^2+2d-4)-2d] \\ & + 3b^2(d-b)^{-2}[d-1-(d-2)\, L(0)] \\ & + 2\sum_{n\neq 0} R_{xx}^3(n)[4-3R_{xx}(n)][1-R_{xx}(n)]^{-2} \\ & + (d-1)\sum_{n} R_{xy}^4(n)[5-3R_{xy}^2(n)][1-R_{xy}^2(n)]^{-2} \end{aligned} \tag{3.11a}$$

where $R_{\alpha\beta}(n) \equiv R_{\alpha\beta}(n, 0)$ is used for convenience of notation. In the 2D

case the factor $[d-1-(d-2)\,L(0)]$ should be replaced by $(1-2/\pi)$ due to (B.4), where the symmetry property $R_{xx}(n_x, n_y) = -R_{xx}(n_y, n_x)$ [see I, Eq. (A4)] enables us to symmetrize the first lattice sum in (3.11a) with respect to interchange of n_x and n_y. The resulting lattice sum is

$$\sum_{n\neq 0} R_{xx}^3(n)[4-3R_{xx}(n)][1-R_{xx}^2(n)]^{-2}$$
$$= \sum_{n\neq 0} R_{xx}^4(n)[5-3R_{xx}^2(n)][1-R_{xx}^2(n)]^{-2} \qquad (3.11b)$$

The final long-time tail for the VACF, exact to $O(c^2)$, again has the form (3.9a), where the coefficient $\beta = \beta(c, \sigma)$, with $\sigma = 1-b$, is now given by

$$\beta(c, \sigma) = c(\pi/2)\, b^2(d-b)^{-2}(d/2\pi)^{1+d/2}(1 + c\beta_1 + \cdots) \qquad (3.12)$$

$O(c^2)$ contributions to τ_0 and τ_1 in (3.7) and (3.9) have not been calculated. The coefficient β_1 involves two lattice sums, which have to be evaluated numerically. This has only been done for the 2D case. The values of $R_{\alpha\beta}(n, 0) = T(0)\, G_{\alpha\beta}(n, 0)$ for several lattice points $n = \{n_x, n_y\}$ are listed in Table I of paper I. The resulting values for $\beta_1(\sigma)$ are listed in Table II of this paper for several values of $b = 1-\sigma$. The above lattice sums converge extremely rapidly, and the contributions from the nearest neighbor sites $\{\pm 1, 0\}$ and $\{0, \pm 1\}$ determine the value of $\beta_1(\sigma)$ with an accuracy of 1.3% over the whole range of σ values.

As mentioned in the introduction, the symmetry relation (1.8) for the VACF under interchange of good and bad conductors and a simultaneous rescaling of time implies the symmetry relation $\beta(c, \sigma) = \beta(1-c, 1/\sigma)$, valid for all d-dimensional simple cubic lattices.

However, for the *square lattice*, there seems to exist an additional sym-

Table II. $O(c^2)$ Expansion Coefficient of $\beta(c, \sigma)$ Defined in (3.12)

σ	$\sigma' = 1/\sigma$	$\beta_1(\sigma) = \beta_1(\sigma')$
0	∞	3.771346
1/10	10	2.026278
1/8	8	1.707141
1/5	5	0.940250
1/4	4	0.551675
1/2	2	−0.538283
2/3	3/2	−0.835464
3/4	4/3	−0.916286
4/5	5/4	−0.949416

metry: $\beta_1(\sigma) = \beta_1(1/\sigma)$, because the coefficient $\beta(c, \sigma)$ in (3.11a) and (3.11b) is invariant under the substitution $\sigma \to \sigma' = 1/\sigma$ or, equivalently, $b \to b' = b/(b-1)$. Note that (3.11a) and (3.11b) for square lattice only depends on b in the combination $[T(0)]^2 = b^2/(2-b)^2$ with $R_{\alpha\beta}(n) = T(0)\, G_{\alpha\beta}(n, 0)$ [see (1.10)]. Thus, the coefficient (3.12) of the long-time tail satisfies the additional symmetry relation

$$\beta(c, \sigma) = \beta(c, 1/\sigma) \tag{3.13}$$

at least to $O(c^2)$ terms included. We conjecture that this symmetry holds for arbitrary values of impurity concentrations and is related to the self-duality of a binary mixture of random resistors (bond problem) on a square lattice. We return to this point in Section 5.

4. BURNETT COEFFICIENTS

The Burnett coefficients are related to the fourth moment of displacement, which is a tensor quantity having two independent coefficients for a lattice with cubic symmetry. We start with the modified Burnett coefficients, defined in (A8):

$$\begin{aligned} \Delta_{xx}(z) &= \tfrac{1}{24} z^2 \{ \langle n_x^4 \rangle(z) - 6z \langle n_x^2 \rangle^2(z) \} \\ \Delta_{xy}(z) &= \tfrac{1}{8} z^2 \{ \langle n_x^2 n_y^2 \rangle(z) - 2z \langle n_x^2 \rangle^2(z) \} \end{aligned} \tag{4.1}$$

Here $\langle n_x^4 \rangle(z)$ and $\langle n_x^2 n_y^2 \rangle(z)$ are Laplace-transformed moments of displacement, which can be obtained from the response or moment-generating function $F(q, z)$ in (1.4) by taking appropriate derivatives with respect to q_x and q_y [see Eq. (A2)].

In paper I the response function was calculated in the form $F(q, z) = g + g^2 M(q, z)$ with $M(q, z)$ given in Eq. (3.6) of paper I and $q = [z + \omega(q)]^{-1}$. One obtains after some algebra

$$\begin{aligned} \Delta_{xx}(z) &= 1/(24d) + \tfrac{1}{24} \{ M^{1111}(0, z) - 6z^{-1} [M^{11}(0, z)]^2 \} \\ \Delta_{xy}(z) &= \tfrac{1}{8} \{ M^{1122}(0, z) - 2z^{-1} [M^{11}(0, z)]^2 \} \end{aligned} \tag{4.2}$$

where superscripts 1 refer to q_x derivatives, and superscripts 2 refer to q_y derivatives, e.g.,

$$M^{1122}(0, z) = [(\partial^4 / \partial q_x^2 \, \partial q_y^2) \, M(q, z)]|_{q=0}$$

Application of (4.2) to $M(q, z)$ yields, after a lengthy, but straightforward, calculation,

$$\Delta_{xx}(z) = \tfrac{1}{12}\Phi(z) - \tfrac{1}{2}c^2 T \sum_n n_x^2 R_{xx}^3(n, z)/[1 - R_{xx}^2(n, z)] \tag{4.3a}$$

$$\Delta_{xy}(z) = -\tfrac{1}{2}c^2 T \sum_n n_y^2 R_{xx}^3(n, z)/[1 - R_{xx}^2(n, z)] - c^2 T \sum_n n_x n_y R_{xy}^3(n, z)/[1 - R_{xy}^2(n, z)] \tag{4.3b}$$

The two-dimensional sums converge at $z = 0$, since the summands decay as $O(|n|^{-4})$, which implies that the modified Burnett coefficients exist for the square lattice. The summand in (4.3a) at $z = 0$ is antisymmetric under interchange of n_x and n_y [see paper I, Eq. (A9)] and we can write the modified Burnett coefficient, exact to $O(c^2)$, as

$$\Delta_{xx}(c, \sigma) \equiv \lim_{z \to 0} \Delta_{xx}(z) = D/12 - c^2 b/[8(2-b)] \sum_n (n_x^2 - n_y^2) R_{xx}^3(n, 0)/[1 - R_{xx}^2(n, 0)] \tag{4.4}$$

The first sum in (4.3b) is the opposite of the one in (4.3a) because of the same antisymmetry. If we denote $R_{xy}^3(n)/[1 - R_{xy}^2(n)] = C(n_x, n_y)$, then $C(n_x, n_y)$ has the same symmetry properties as $G_{xy}(n_x, n_y)$ in Eqs. (A5) and (A6) of paper I, and symmetrization of the summand yields the second modified Burnett coefficient, exact to $O(c^2)$:

$$\Delta_{xy}(c, \sigma) \equiv \Delta_{xy}(z = 0) = -(\Delta_{xx} - D/12) - c^2 b/[8(2-b)] \times \sum_n (2n_x + 1)(2n_y - 1) R_{xy}^3(n, 0)[1 - R_{xy}^2(n, 0)] \tag{4.5}$$

The summands in (4.4) and (4.5) have the same symmetry properties as $H(n_x, n_y)$ in Eq. (4.8) of paper I. The integrals $R_{\alpha\beta}(n, 0) = \frac{1}{2}[b/(2-b)]\, G_{\alpha\beta}(n, 0)$ are listed in Table 1 of paper I for several lattice sites $n = (n_x, n_y)$ near the origin. The total tail contribution originating from all terms with $|n| > N$ decays here very slowly, i.e., $O(N^{-2})$ as $N \to \infty$, whereas the total contribution of the lattice sum in Section 3 decays as $O(N^{-6})$. The resulting values of the coefficients are listed in Table III for several values of $\sigma = 1 - b$. The symmetry of the average random lattice under the interchange of good and bad conductances leads to the same symmetry relation as for the diffusion coefficient:

$$\Delta_{xx}(c, \sigma) = \sigma \Delta_{xx}(1 - c, 1/\sigma)$$
$$\Delta_{xy}(c, \sigma) = \sigma \Delta_{xy}(1 - c, 1/\sigma) \tag{4.6}$$

Table III. Burnett Coefficients, Defined in (4.4) and (4.7)

σ	$\sigma' = 1/\sigma$	$c^{-2}\Delta'_{xx}$	$c^{-2}\Delta_{xy}$
0	∞	1.350 E−2	6.268 E−3
1/10	10	5.920 E−3	2.653 E−3
1/8	8	4.814 E−3	2.144 E−3
1/5	5	2.572 E−3	1.127 E−3
1/4	4	1.679 E−3	7.295 E−4
1/2	2	1.574 E−4	6.68 E−5
2/3	3/2	2.03 E−5	8.6 E−6
3/4	4/3	5.3 E−6	2.2 E−6
4/5	5/4	1.9 E−6	8.1 E−7

We further note that the lattice sums in (4.4) and (4.5) are invariant under the transformation $\sigma = 1 - b \to \sigma' = 1/\sigma$ or $b \to b' = b(b-1)$ because they only depend on b in the invariant combination $[T(0)]^2 \sim b^2/(2-b)^2$. Thus, the Burnett coefficients in the density-expanded forms (4.4) and (4.5) have the additional symmetry

$$\begin{aligned} \Delta'_{xx}(c, \sigma) &= \Delta'_{xx}(c, 1/\sigma) \\ \Delta_{xy}(c, \sigma) &= \Delta_{xy}(c, 1/\sigma) \end{aligned} \tag{4.7}$$

where $\Delta'_{xx} = \Delta_{xx} - D/12$. This symmetry is also present in the coefficient $\beta(c, \sigma)$ of the long-time tail of the VACF and is conjectured to be related to the self-duality of bond percolation on a square lattice.

If one is interested in the dominant small-z singularity of the modified Burnett coefficients, one can show that $\delta\Delta_{xy}(z) \simeq O(z \ln z)$, using a similar method as in (3.1). If one applies the same method to $\Delta_{xx}(z)$, one finds that the coefficient of $O(z \ln z)$ diverges logarithmically, suggesting that $\delta\Delta_{xx}(z) \sim O[z(\ln z)^2]$.[2] However, we have not analyzed the dominant singularity in $\Delta_{\alpha\beta}(z)$ and its corresponding long-time tail any further, since the ordinary Burnett coefficients have a longer time tail, which is entirely determined by the tail of the VACF $\phi(t) \simeq -at^{-2}$, as we shall see below.

The ordinary Burnett coefficients, defined in (1.5) for an isotropic system and in (A5) for a system with cubic symmetry, are related to the long-time behavior of the fourth cumulant of the displacement. This behavior can be inferred from (A11), using

$$\int_0^\infty dt\, \tilde{\Delta}_{\alpha\beta}(t) = \lim_{z \to 0} \Delta_{\alpha\beta}(z) \equiv \Delta_{\alpha\beta}(c, \sigma) \tag{4.8}$$

[2] J. M. Luck and Th. M. Nieuwenhuizen (private communication) have proved this conjecture by showing that the small-z singularity of $\sum n_x^2 G_{xx}^3(n, z)$ and $\sum n_y^2 G_{xx}^3(n, z)$ equals $(3/\pi^2)(\log z)^2$.

where $\tilde{\Delta}_{\alpha\beta}(t)$ is the inverse Laplace transform of $\Delta_{\alpha\beta}(z)$ and $\Delta_{\alpha\beta}(c, \sigma)$ as given in (4.4) and (4.5), with values listed in Table III. We further used the long-time behavior of the VACF $\phi(t) \simeq -\beta/t^2$ in (3.12) to show

$$\tilde{D}(t) = \int_0^t d\tau\, \phi(\tau) \simeq D + \beta/t, \qquad t \to \infty$$
$$\int_0^t d\tau\, \tilde{D}(\tau)[\tilde{D}(t) - \tilde{D}(t-\tau)] \simeq -\beta D \ln t + \text{const}, \qquad t \to \infty \tag{4.9}$$

Thus, we deduce from (A11) that the long-time behavior of fourth and second moments are related through

$$\lim_{t\to\infty} \left\{ \tilde{B}_{xx}(t) + \int_0^t d\tau\, \tilde{D}(\tau)[\tilde{D}(t) - \tilde{D}(t-\tau)] \right\} \tag{4.10a}$$
$$= \lim_{t\to\infty} [\tilde{B}_{xx}(t) - \beta D \ln t] = \Delta_{xx}(c, \sigma) \tag{4.10b}$$

and

$$\lim_{t\to\infty} [\tilde{B}_{xx}(t) - \tilde{B}_{xy}(t)] = \Delta_{xx}(c, \sigma) - \Delta_{xy}(c, \sigma) \tag{4.11}$$

The above limits exist and their values are listed in Table III.

From the above results one can derive some interesting conclusions: First, we observe that the ordinary Burnett coefficient, defined as the long-time limit of $\tilde{B}_{\alpha\beta}(t)$ in (A6), does not exist for the RBM on the square lattice, since $\tilde{B}_{\alpha\beta}(t) \sim \ln t$ as $t \to \infty$. The approach of $B_{xx}(t)$ to its asymptotic form in (4.10b), $[\beta D \ln t + \Delta_{xx}(c, \sigma)]$, is expected to be $O(t^{-1} \ln t)$ and has been confirmed (see footnote 2). However, for $d \geqslant 3$ the Burnett coefficient exists. This is the standard behavior for Lorentz-type models,[17,18] but here also a subleading asymptotic correction $\Delta(c, \sigma)$ has been calculated. However, the modified Burnett coefficient defined through (4.1) and (4.8) does exist for the RBM with $d \geqslant 2$, at least to $O(c^2)$ terms included. The latter result confirms a conjecture by Alley and Alder[14] on the existence of the modified Burnett coefficient. In the case of fluids the conjecture has been disproved.[16] Our results further suggest that Eq. (4.10a) would be the appropriate way to correlate the long-time behavior of $\langle n_x^4 \rangle$ and $\langle n_x^2 \rangle$ in future computer simulations.

Finally, Eq. (4.11) shows that the divergent terms in $\tilde{B}_{xx}(t)$ and $\tilde{B}_{xy}(t)$ at times cancel. This result is to be expected on the basis of isotropic Lorentz models, where $\tilde{B}_{xx}(t) \equiv \tilde{B}_{xy}(t)$ at all times. The reason is that the dynamic events contributing to long-time tails come from *large-scale* hydrodynamic fluctuations in which our *lattice model* can be considered as *continuous* and *isotropic*.

5. DISCUSSION

This paper studies the random barrier model (RBM) or random resistor network on a hypercubic lattice with a concentration $c = 1 - p$ of impurity bonds (with conductance $\sigma = 1 - b$) and a concentration $p = 1 - c$ of host lattice bonds (with conductance $\sigma_0 = 1$). Bond percolation, or the "ants in a labyrinth" model, corresponds to the parameter value $\sigma \equiv 1 - b = 0$. For $\sigma \gg 1$ one has superconducting bonds or the termite model.

All results are obtained in the form of expansions in powers of $c = 1 - p$. In the percolation literature such results are referred to as "high-density expansions" [4]. Virtually *no such expansion coefficients are known* for d.c. and a.c. transport coefficients. "High density" or c-expansions have only been given for the percolation probability and some moments of the cluster size distribution.[5] On the other hand, the coefficients in the "low-density" or p-expansion are known to rather high order.[6]

The paper concentrates on the VACF $\phi(t)$ and the time-dependent diffusion coefficient $\tilde{D}(t)$, defined as

$$\begin{aligned} \phi(t) &= D_E \delta_+(t) + \phi_+(t) \\ \tilde{D}(t) &= D_E + \int_0^t d\tau\, \phi_+(\tau) \end{aligned} \tag{5.1}$$

The corresponding results for the Burnett functions are briefly summarized in Section 4. The most important new results are the *long-time tails* of (5.1), exact to $O(c^2)$, and the *complete time dependence* of these quantities, exact to $O(c)$. We first discuss the last results, which are given through (2.7), (2.13), and (2.22).

The most striking property of the VACF is the *cage effect*, referring to the existance of memory effects that lead to negative velocity correlations. Here $\phi_+(t)$ is *negative* for all $t > 0$. The cage effect results from a single or repeated returns ("ring collisions" in kinetic theory jargon) of the random walker (RW) with the same impurity. The hoppings on the host lattice are the analogs of "uncorrelated binary collisions" of the moving particle in a deterministic Lorentz gas. The present hopping models on disordered lattices are referred to as stochastic Lorentz models.[32]

Regarding $\tilde{D}(t)$, we note that the cage effect with its negative velocity correlations substantially reduces the high-frequency or Enskog diffusion coefficient $D_E = \tilde{D}(t \downarrow 0)$, so that the ordinary long-time diffusion coefficient $D(c, \sigma) = \tilde{D}(t \to \infty)$ is much smaller than $D_E = D_0(1 - bc)$. The latter coefficient describes diffusion on an effective uniform lattice with an effective number of sites $N(1 - bc)$.

The short-time behavior of VACF and $\tilde{D}(t)$ at *arbitrary density* was

fully discussed in Ref. 3. The intermediate-time behavior of VACF in $O(c)$ requires a Laplace inversion [see (2.7)]. For poorly conducting impurities ($0 \leqslant \sigma < 1$ or $0 < b \leqslant 1$) the inverse Laplace transform is completely determined by the contribution $\phi_b(t)$ from a branch cut. For the square lattice it is given by (2.21) and evaluated numerically for several values of $b = 1 - \sigma$. For the value $b = 1$ (percolation, ants in a labyrinth) the resulting VACF is plotted in Fig. 1 for both short and long times (see insert, where $1 \leqslant t \leqslant 100$). The corresponding behavior of the reduced diffusion coefficient $d(t)$ defined in (2.22) is shown as a function of $1/t$ in Fig. 4.

Different behavior is found if the impurity bonds have higher conductivity than the host lattice bonds ($\sigma > 1$ or $b < 0$). Then there is, apart from the above $\phi_b(t)$, also a contribution $\phi_p(t)$ from a pole, provided $\sigma > 1$ for $d \leqslant 2$ and $\sigma > \sigma_0(d) = 1 + d/[2L(0) - 1]$ for the hypercubic lattice with $d > 2$ [see below (2.15)]. For $\sigma \gg 1$ (termite model, superconducting impurities) or for σ slightly above σ_0 the location of the pole z_p can be determined analytically [see (2.16) and (2.17)] by solving (2.8). For general σ values the root z_p of (2.8) has to be determined numerically. The contribution $\phi_p(t)$ for the square lattice is explicitly given by (2.20). Combining the numerical values for (2.20) with those for $\phi_b(t)$ in (2.21) yields the result for the VACF to $O(c)$ plotted in Fig. 2 ($\sigma = 1 - b = 2$) and Fig. 3 ($\sigma = 1 - b = 11$).

For large σ (see Fig. 3) the VACF shows interesting structure already in $O(c)$. On the short time scale of $1/\sigma$ seconds the termite hops essentially back and forth across an isolated superconducting bond or sits at one of its two ends. There is only a small probability, proportional to $1/\sigma$ per hop, of wandering off into the normal conducting region. This behavior is described by the exponential contribution (2.16) from the pole (see insert Fig. 3, solid curve labeled b), which essentially describes the complete VACF (solid curve, labeled a) for $t < 1$. Note that the time scale ($0.01 \leqslant t \leqslant 100$) on the insert in Fig. 3 differs from that on the inserts in Figs. 1 and 2. The corresponding reduced time-dependent diffusion coefficient $d(t)$ is shown in Fig. 6. It is negative because of the negative factor b in the normalization on the rhs of (2.22). Figures 2 and 5 show the VACF and diffusion coefficient at an intermediate σ value ($\sigma = 1 - b = 2$).

The long-time behavior of the VACF, $\phi(t) \simeq -\beta(c, \sigma)\, t^{-1-d/2}$ in (3.7), and of the time-dependent diffusion coefficient

$$\tilde{D}(t) \simeq D(c, \sigma) + [2\beta(c, \sigma)/d]\, t^{-d/2}$$

is calculated exactly to $O(c^2)$ for the square lattice with $\beta(c, \sigma)$ given in (3.12) in combination with Table II. Calculation of the coefficient $\beta(c, \sigma)$ in (3.10) and (3.11) to $O(c^2)$ requires the evaluation of lattice sums describing

all possible visits of the RW to two different impurities. The sums, which converge rapidly, have been evaluated numerically. The present RBM together with the related hopping model on a square lattice with excluded sites[1] is the only model for which the dominant long-time tail has been calculated to $O(c^2)$ in the impurity concentration.

The coefficient of the tail for the RBM exhibits an unexpected symmetry $\beta(c, \sigma) = \beta(c, 1/\sigma)$, valid for the self-dual square lattice only, as verified below (3.12) to $O(c^2)$. It is conjectured[2] that this symmetry holds for arbitrary impurity concentration and is a consequence of the self-duality of the RBM on the square lattice. This conjecture is based on (i) mode coupling[18] and kinetic theory[33] calculations, predicting that $\phi(t) \sim -\langle(\delta D)^2\rangle\, t^{-1-d/2}$ as $t \to \infty$, where $\langle(\delta D)^2\rangle$ is the local fluctuation in the spatially varying diffusion coefficient of the disordered system, and (ii) the work of Wright *et al.*,[34] who used self-duality arguments to show that the local fluctuations in the resistivity, which are proportional to $\langle(\delta D)^2\rangle$, satisfy the same symmetry.

The long-time tails are caused by the repeated return of the RW to the same impurity ("repeated ring collisions"), which is the same mechanism causing long-time tails in fluids and stationary random media.[14–18] At low densities there is an interesting difference between a deterministic Lorentz gas, where $D(c) \sim 1/c$ as $c \to 0$, and a hopping model, where $D(c) \sim \text{const}$ as $c \to 0$. In the former model the coefficient $\beta(c, \sigma)$ for $c \to 0$ is only determined by the ring collisions,[16,17] whereas in the latter models one has to sum all repeated ring collisions. If $\phi_R(t)$ and $\phi_{RR}(t)$ denote, respectively, the VACF at long times obtained from the ring (R) and repeated ring (RR) collisions, one has for the present hopping model

$$\phi_{RR}(t) \simeq [d^2/(d-b)^2]\, \phi_R(t), \qquad t \to \infty \tag{5.2}$$

There have been many unsuccessful attempts to test the results on long-time tails in Lorentz gases obtained from computer simulations against those from theoretical predictions, as discussed in Ref. 18. The present model offers a promising candidate for a successful comparison because we have also determined the first subleading correction to the tail, which is of the form

$$\phi(t) = \begin{cases} -\beta t^{-1-d/2}[1 + \tau_0/t + O(t^{-2})], & d > 2 \\ -\beta t^{-2}[1 + (\tau_0/t)\ln(t/\tau_1) + O(t^{-2})], & d = 2 \end{cases} \tag{5.3}$$

and the coefficients τ_0 and τ_1 have been calculated in (3.9) and (3.7) to lowest nonvanishing order in c.

To illustrate the importance of the subleading terms in the VACF to

$O(c)$ for the two-dimensional case, we have plotted in Figs. 1–3 (dashed lines at large t) both the dominant tail of $O(t^{-2})$ as well as the tail with the correction term included. The latter form (dashed lines at large t, Figs. 1–3) essentially coincides with the exact $O(c)-$VACF (solid lines) for $t \gtrsim 5$. The inserts in Fig. 1 ($\sigma=0$) and Fig. 3 ($\sigma=11$) clearly show for the present model that one cannot see the pure t^{-2} tail in the time interval between 10 and 60 mean hopping ("collision") times.[1-3] Unfortunately, this time interval covers the typical range where one has searched for the long-time tail of the VACF in computer simulations on the Lorentz gas.[14,15,18] This suggests that a detailed comparison of computer simulations and kinetic theory calculations for Lorentz-type models is only feasible for the time intervals presently accessible in computer simulations if at least the next asymptotic correction is known theoretically. This has been shown convincingly in recent computer simulations by Frenkel[19] on the hopping model on a square lattice with randomly excluded sites.[1]

Next, it is instructive to consider the long-time behavior of the diffusion coefficient $\tilde{D}(t)$ in (5.1), for which the same information can be obtained as for VACF. In Figs. 4–6 the reduced diffusion coefficient $d(t)$ defined in (2.22) is plotted as a function of $1/t$ (solid lines) for the square lattice and compared with the dominant tail and with the first correction (3.8) included (dashed curves, left side of graphs). The latter curve is very close to the actual VACF; the first one is not, except for $\sigma=2$ (Fig. 5), where *all* differences at long times are rather small.

In connection with the poor agreement between computer simulations and kinetic theory calculations in the Lorentz gas we compare it with our results for the percolation model (RBM with $\sigma=1-b=0$). Suppose the solid curve in Fig. 4 would represent a smooth fit to the simulation results. If one were to fit this curve in the interval, say, between 10 and 60 mean hopping ("collision") times with a linear curve $\tilde{D}(t)=D(c)+\beta(c)(t^{-1})$, one would find an **apparent slope** β_{sim} which would be 20–30% larger than the **actual slope** $\beta(c)$ of the dominant tail (see upper dashed line in Fig. 4). This observation may partially explain why the "apparent" β values reported from computer simulations on the Lorentz gas[14,15] are too large.

Next, we comment about the termite limit ($\sigma \to \infty$). If one performs the c-expansion first [as is done in (1.9)], and subsequently takes the limit of the coefficient as $\sigma \to \infty$ or $b \to -\infty$, the scattering function in (1.10a) approaches a finite limit $T(z)=-[2dJ(z)]^{-1}$ and all coefficients in (1.9) remain finite. However, the proper procedure for the termite model would be to take first $\sigma \to \infty$ and subsequently perform a c expansion. The expression for the transition rate (1.2) suggests that both limits cannot be interchanged, so that the expansions derived here can only be applied to $\sigma=1-b \gg 1$ as long as $|b|\, c \simeq \sigma c \ll 1$.

We finally compare our exact results for the VACF with the results of the effective medium approximation (EMA). This approximation takes into account all terms containing less than four $T(z)$ operators [27], i.e., to $O(c^2)$ all terms in (1.9) except the two lattice sums. Extension of the results in Refs. 9 and 10 yields the long-time behavior of the VACF in the form (3.7a), i.e.,

$$\phi(t) \simeq -\beta t^{-2}[1 + (\tau_0/t)\ln(t/\tau_1) + \cdots] \tag{5.4}$$

For the percolation case ($\sigma = 1 - b = 0$) the EMA values of these coefficients are

$$\begin{aligned} \beta(c) &= (c/2\pi)(1-2c)^{-1} \\ \tau_0(c) &= (4/\pi)(1-2c)^{-1} \\ \tau_1(c) &= (1/8)(1-2c)^{-1}\exp[3/2 - \gamma + c - (\pi/4)(1-2c)] \end{aligned} \tag{5.5}$$

The EMA value for the diffusion coefficient is $D(c) = (1-2c)/4$ and vanishes at the percolation threshold $c = 1/2$. The EMA values (5.5) are exact to $O(c)$ [compare with (3.7b)]. However, the EMA value for β_1 defined in (3.10) equals $\beta_1(\text{EMA}) = 2$ [see (5.5)] and differs from the exact value $\beta_1 \simeq 3.77$ (see Table II). As $c \uparrow 1/2$ (threshold), both coefficients $\beta(c)$ and $\tau_0(c)$ are divergent, and the long-time behavior is dominated by the stronger tail[10]

$$\phi(t) \simeq -(4\pi\sqrt{2})^{-1}(\ln t)^{1/2}\, t^{-3/2} \tag{5.6}$$

The last remark may explain the observation,[14] made in computer simulations on the overlapping Lorentz gas, where the tail of the VACF is fitted to $\phi_{\text{sim}}(t) \simeq -At^{-\alpha}$. There it is observed that the "apparent" exponent α decreases from its low-density value $\alpha \simeq 2$ to $\alpha \simeq 1$ as the density of scatterers approaches the continuum percolation threshold.

APPENDIX A

In this Appendix we derive the relations connecting response function, moments of displacements, diffusion and Burnett coefficients, and VACF for a system with cubic symmetry.

The response function $F(q, z)$ defined in (1.4) generates the Laplace-transformed moments of displacements,

$$F(q, z) = 1/z - \tfrac{1}{2}q^2\langle(\hat{q}\cdot n)^2\rangle(z) + \tfrac{1}{24}q^4\langle(\hat{q}\cdot n)^4\rangle(z) + \cdots \tag{A1}$$

where $\hat{q} \cdot n = \sum \hat{q}_\alpha n_\alpha$ with $n = \{n_a\}$ and $q = \{q_\alpha\}$ being d-dimensional vectors ($\alpha = x, y, \ldots, d$), and $\hat{q}$ is a unit vector parallel to q. Thus

$$\langle n_\alpha n_\beta \cdots \rangle(z) = \left[\frac{\partial}{\partial i q_\alpha} \frac{\partial}{\partial i q_\beta} F(q, z) \right]_{q=0} \tag{A2}$$

For a system with cubic symmetry, $\langle n_\alpha n_\beta \rangle = \delta_{\alpha\beta} \langle n_x^2 \rangle$ is a symmetric, second-rank tensor with one independent nonvanishing element. Similarly, $\langle n_\alpha n_\beta n_\gamma n_\delta \rangle$ is a fourth-rank tensor with only two independent elements, say $\langle n_x^4 \rangle$ and $\langle n_x^2 n_y^2 \rangle$, so that

$$\begin{aligned} \langle (\hat{q} \cdot n)^2 \rangle &= \langle n_x^2 \rangle \sum_\alpha \hat{q}_\alpha^2 \\ \langle (\hat{q} \cdot n)^4 \rangle &= \langle n_x^4 \rangle \sum_\alpha \hat{q}_\alpha^4 + 3 \langle n_x^2 n_y^2 \rangle \sum_{\alpha \neq \beta} \sum \hat{q}_\alpha^2 \hat{q}_\beta^2 \end{aligned} \tag{A3}$$

Let $\tilde{F}(q, t)$ be the Fourier transform of the average probability $\langle p(nt; m0) \rangle$. Burnett coefficients can be introduced by extending Fick's law with $O(\nabla^4)$ terms, which reads, after Fourier transformation,

$$(\partial/\partial t)\, \tilde{F}(q, t) = [-q^2 D(\hat{q}) + q^4 B(\hat{q}) + \cdots]\, \tilde{F}(q, t) \tag{A4}$$

The diffusion coefficient $D(\hat{q})$ and Burnett coefficient $B(\hat{q})$ are second- and fourth-rank tensors with cubic symmetry. They can be related to $(\partial/\partial t) \ln \tilde{F}(q, t)$, where $\ln \tilde{F}(q, t)$ generates the cumulants of displacements:

$$\begin{aligned} \tilde{D}(\hat{q}, t) &= \frac{1}{2} \left(\frac{\partial}{\partial t} \right) \langle (\hat{q} \cdot n)^2 \rangle = \tilde{D}(t) \\ \tilde{B}(\hat{q}, t) &= \frac{1}{24} \left(\frac{\partial}{\partial t} \right) \{ \langle (\hat{q} \cdot n)^4 \rangle - 3 \langle (\hat{q} \cdot n)^2 \rangle^2 \} \\ &= \tilde{B}_{xx}(t) \sum_\alpha \hat{q}_\alpha^4 + \tilde{B}_{xy}(t) \sum_{\alpha \neq \beta} \sum \hat{q}_\alpha^2 \hat{q}_\beta^2 \end{aligned} \tag{A5}$$

with

$$\begin{aligned} \tilde{B}_{xx}(t) &= \frac{1}{24} \left(\frac{\partial}{\partial t} \right) (\langle n_x^4 \rangle - 3 \langle n_x^2 \rangle^2) \\ \tilde{B}_{xy}(t) &= \frac{1}{8} \left(\frac{\partial}{\partial t} \right) (\langle n_x^2 n_y^2 \rangle - \langle n_x^2 \rangle^2) \end{aligned} \tag{A6}$$

The diffusion and Burnett coefficients can be identified (if they exist) with $D = \lim_{t \to \infty} \tilde{D}(t)$ and $B_{\alpha\beta} = \lim_{t \to \infty} \tilde{B}_{\alpha\beta}(t)$. In the ordinary Lorentz gas,

which has isotropic instead of cubic symmetry, there is only one independent Burnett coefficient, $\tilde{B}_{xx}(t) = \tilde{B}_{xy}(t)$.

In generalized hydrodynamics one defines frequency-dependent transport coefficients through the response functions[14,16] as

$$F(q, z) = [z + q^2 \Phi(\hat{q}, z) - q^4 \Delta(\hat{q}, z) + \cdots]^{-1} \tag{A7}$$

By making a q-expansion and comparing coefficients with (A1), one finds the frequency-dependent diffusion coefficient

$$\Phi(\hat{q}, z) = \Phi(z) = \tfrac{1}{2} z^2 \langle (\hat{q} \cdot n)^2 \rangle(z) = \tfrac{1}{2} z^2 \langle n_x^2 \rangle(z) \tag{A8}$$

where the static diffusion coefficient is $D = \Phi(z = 0)$. Similarly, one has

$$\begin{aligned} \Delta(\hat{q}, z) &= \tfrac{1}{24} z^2 [\langle (\hat{q} \cdot n)^4 \rangle(z) - 6z \langle (\hat{q} \cdot n)^2 \rangle^2(z)] \\ &= \Delta_{xx}(z) \sum_{\alpha} \hat{q}_\alpha^4 + \Delta_{xy}(z) \sum_{\alpha} \sum_{\alpha \neq \beta} \hat{q}_\alpha^2 \hat{q}_\beta^2 \end{aligned} \tag{A9}$$

with

$$\begin{aligned} \Delta_{xx}(z) &= \tfrac{1}{24} z^2 [\langle n_x^4 \rangle(z) - 6z \langle n_x^2 \rangle^2(z)] \\ \Delta_{xy}(z) &= \tfrac{1}{8} z^2 [\langle n_x^2 n_y^2 \rangle(z) - 2z \langle n_x^2 \rangle^2(z)] \end{aligned} \tag{A10}$$

Here $\Delta_{\alpha\beta} = \lim_{z \to 0} \Delta_{\alpha\beta}(z)$ are the modified Burnett coefficients[14,16] if the limits exist. We further observe that the inverse Laplace transform of (A8) can be identified as the lattice equivalent of the velocity autocorrelation function (VACF):

$$\phi(t) = \mathscr{L}_t^{-1}(\Phi(z)) = \tfrac{1}{2} (\partial/\partial t)^2 \langle n_x^2 \rangle = \langle v_x(t)\, v_x(0) \rangle \tag{A11}$$

Similarly $\mathscr{L}_t^{-1}(\Delta(z))$ and $B(t)$ can be expressed in four-point velocity correlation functions,[16] but these explicit expressions are not of interest here.

By comparing (A5), (A6), and (A9) one obtains the following relations:

$$\begin{aligned} \int_0^t d\tau\, \tilde{\Delta}_{xy}(\tau) &= \tilde{B}_{xy}(t) + \int_0^t d\tau\, \tilde{D}(\tau)[\tilde{D}(t) - \tilde{D}(t - \tau)] \\ \int_0^t d\tau\, \tilde{\Delta}_{xy}(\tau) &= \tilde{B}_{xy}(t) + \int_0^t d\tau\, \tilde{D}(\tau)[\tilde{D}(t) - \tilde{D}(t - \tau)] \end{aligned} \tag{A12}$$

where $\tilde{\Delta}_{\alpha\beta}(t) = \mathscr{L}_t^{-1}(\Delta_{\alpha\beta}(z))$. If the long-time limit of the lhs of (A11) exists, then it approaches $\lim_{z \to 0} \Delta_{\alpha\beta}(z) = \Delta_{\alpha\beta}$.

APPENDIX B

In the evaluation of the long-time tail in Section 3.2 one needs to calculate the lattice sum

$$\sum_{n\neq 0} R_{xx}^2(n, z) = 4T^2(z)\left\{\int_q g^2(1-\cos q_x)^2 - \left[\int_q g(1-\cos q_x)\right]^2\right\} \tag{B1}$$

with $g = 1/[z+\omega(q)]$. Here we have used (1.8) and (1.9) and the relation

$$\sum_{n\neq 0} e^{-in(q+q')} = (2\pi)^d\,\delta(q+q') - 1 \tag{B2}$$

The second integral in (B1) yields

$$\int g(1-\cos q_x) = dJ(z) = 1 - zL(z)$$

due to (1.8), (1.9), and (2.2). The first integral can be further reduced to

$$\begin{aligned}\int g^2(1-\cos q_x)^2 &= 2\int g^2\omega(q) - \int g^2 \sin^2 q_x \\ &= -2dJ'(z) + d^2J(z) - dL(z)\end{aligned}$$

In the second equality we have performed a partial integration using the relation

$$(d/dq_x)[z+\omega(q)]^{-1} = -d^{-1}\sin q_x/[z+\omega(q)]^2$$

Combination of the previous equations yields finally

$$\sum_{n\neq 0} R_{xx}^2(n, z) = 4dT^2(z)[dJ(z) - dJ^2(z) - 2J'(z) - L(z)] \tag{B3}$$

In fact we need the value of (B3) only for $z \to 0$, yielding

$$\sum_{n\neq 0} R_{xx}^2(n, 0) = b^2(d-b)^{-2}\begin{cases}[d-1-(d-2)\,L(0)], & d\geqslant 3\\ (1-2\pi), & d=2\end{cases} \tag{B4}$$

For $d=2$ we have used the relation $L(z) \simeq -\pi^{-1}\ln z$ as $z \to 0$.

In principle one could calculate higher order lattice sums, such as $\sum R^3(n, z)$, using the analog of (B2):

$$\sum_{n\neq 0} \exp[in(q_1+q_2+q_3)] = (2\pi)^d\,\delta(q_1+q_2+q_3) - 1$$

This relation couples, however, the integration variables q_1, q_2, q_3 in the different q-integrations and does not lead to any simplifications.

ACKNOWLEDGMENTS

The author is grateful to Dr. Peter van Velthoven for performing the numerical work reported in this paper. He further thanks the Physics Department of the University of Florida for its hospitality during his stay from November 1986 to April 1987. This research was supported in part by National Science Foundation grant CHE-8411932.

REFERENCES

1. Th. M. Nieuwenhuizen, P. F. J. van Velthoven, and M. H. Ernst, *Phys. Rev. Lett.* **51**:2766 (1986).
2. M. H. Ernst, P. F. J. van Velthoven, and Th. M. Nieuwenhuizen, *J. Phys. A: Math. Gen.* **20**:947 (1986).
3. M. H. Ernst and P. F. J. van Velthoven, *J. Stat. Phys.* **45**:1001 (1986).
4. J. W. Haus and K. W. Kehr, *Phys. Rep.* (1987), to appear.
5. M. F. Sykes and M. K. Wilkinson, *J. Phys. A: Math. Gen.* **19**:3415 (1986).
6. R. Fish and A. B. Harris, *Phys. Rev. B* **18**:416 (1978); J. W. Essams and F. M. Bhatti, *J. Phys. A: Math. Gen.* **18**:3577 (1985); J. Adler, *J. Phys. A: Math. Gen.* **18**:307 (1985).
7. F. Wu, *Rev. Mod. Phys.* **54**:235 (1982).
8. S. Kirkpatrick, *Rev. Mod. Phys.* **45**:574 (1973).
9. T. Odagaki, M. Lax, and A. Puri, *Phys. Rev. B* **28**:2755 (1983).
10. M. Sahimi, B. D. Hughes, L. E. Scriven, and H. T. Davis, *J. Chem. Phys.* **78**:6849 (1983).
11. T. Odagaki, *Phys. Rev. B* **33**:544 (1986).
12. U. M. S. Costa, C. Tsallis, and G. Schwachheim, *Phys. Rev. B* **33**:510 (1986).
13. M. Sahimi and H. Siddiqui, *J. Phys. A: Math. Gen.* **18**:L-727 (1985); F. Leyvzaz, J. Adler, A. Aharony, A. Bunde, A. Coniglio, D. C. Hong, H. E. Stanley, and D. Stauffer, *J. Phys. A: Math. Gen.* **19**:3683 (1986).
14. B. J. Alder and W. E. Alley, *J. Stat. Phys.* **19**:341 (1978).
15. B. J. Alder and W. E. Alley, *Physica* **121A**:523 (1983); C. Bruin, *Physica* **72**:261 (1974); J. C. Lewis and J. A. Tjon, *Phys. Lett.* **66A**:349 (1978).
16. M. H. Ernst and A. Weyland, *Phys. Lett.* **34A**:39 (1971).
17. M. H. Ernst and H. van Beijeren, *J. Stat. Phys.* **26**:1 (1981).
18. J. Machta, M. H. Ernst, H. van Beijeren, and J. R. Dorfman, *J. Stat. Phys.* **35**:413 (1984).
19. D. Frenkel, *Phys. Lett. A* **121**:385 (1987).
20. T. Keyes and J. W. Lyklema, *J. Stat. Phys.* **27**:487 (1982).
21. J. J. Brey, J. Gomez Ordonez, and A. Santos, preprint (1987).
22. D. C. Hong, H. E. Stanley, A. Coniglio, and A. Bunde, *Phys. Rev. B* **33**:4564 (1986).
23. J. R. Dorfman and H. van Beijeren, in *Statistical Mechanics, Part B, Time Dependent Processes*, B. J. Berne, ed. (Plenum Press, New York, 1977).
24. B. J. Alder and T. E. Wainwright, *Phys. Rev. A* **1**:18 (1970); E. G. D. Cohen, *Phys. Today* (January):64 (1984).
25. E. H. Hauge, in *Transport Phenomena*, G. Kirczenow and J. Marro, eds. (Lecture Notes in Physics, No. 31, Springer-Verlag, Berlin, 1974), p. 338; E. G. D. Cohen, Colloques Internationaux CNRS No. 236—Théories Cinétiques Classiques et Relativistes.

26. I. M. de Schepper and M. H. Ernst, *Physica* **93A**:611 (1978); **98A**:189 (1979); M. H. Ernst and J. R. Dorfman, *J. Stat. Phys.* **15**:7 (1976).
27. P. J. H. Denteneer and M. H. Ernst, *J. Phys. C Solid State Phys.* **C16**:L-961 (1983).
28. P. J. H. Denteneer and M. H. Ernst, *Phys. Rev. B* **29**:1755 (1984).
29. Th. M. Nieuwenhuizen and M. H. Ernst, *Phys. Rev. B* **31**:3518 (1985).
30. G. L. Montet, *Phys. Rev. B* **7**:650 (1972).
31. M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1972).
32. H. van Beijeren, *Rev. Mod. Phys.* **54**:195 (1982).
33. A. K. Harrison and R. Zwanzig, *J. Stat. Phys.* **42**:935 (1986).
34. D. C. Wright, D. J. Bergman, and Y. Kantor, *Phys. Rev. B* **33**:396 (1986).