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Transport anisotropy and percolation in the two-dimensional random-hopping model

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We consider hopping transport on an anisotropic two-dimensional square lattice. The displacements parallel to one axis are governed by uniform, nearest-neighbor hopping rates c, while the displacements parallel to the other axis are governed by static but spatially fluctuating rates w_n. Adapting a new class of generating functions recently introduced for the random-trapping problem, we are able to obtain expressions for the mean-square displacement in the fluctuating direction through an exact decoupling of the effects due to displacements in the uniform direction. The resulting expressions for the low-frequency diffusion coefficient $D(\epsilon)$ are exact in the limits $c\rightarrow 0$ $[D(0)=\langle 1/w\rangle^{-1}]$ and $c\rightarrow\infty$ $[D(0)=\langle w\rangle]$. Moreover, when the condition of long-time isotropy is imposed we obtain expressions which are, to lowest order in the fluctuations, identical to results obtained in the effective-medium approximation for the square lattice with fluctuating rates in both directions. The present method offers the possibility of systematic improvements to the effective-medium results for the dc conductivity and frequency corrections.

I. INTRODUCTION

In many amorphous materials hopping transport between localized states is the basic mechanism underlying electronic conduction, the incoherent migration of spin states, and the transport of excited electronic and vibrational energy.[1–16] Accordingly, several simple but insightful models have been developed to understand such phenomena. The *random-hopping* model is one such model in which a particle jumps between localized states that are separated by random barriers of varying heights. The randomness in the barrier heights give rise to a distribution of symmetric hopping or jump rates which, typically, are assumed to be independent stochastic variables governed by a single probability distribution function.[1,5–12] In this paper we focus on the random-hopping problem in two dimensions.

It is important at this point to distinguish the random-hopping model from the *random-trapping* model,[5,13–16] in which the particle performs a series of uncorrelated jumps on an ordered array of random wells of varying depths. It is now the randomness in *well depths* that produces a distribution of jump rates. The symmetry condition relating forward and backward jumps is different, however, from that which is imposed in the random-hopping model. Alexander[5] has shown that the two models display significant qualitative differences for dimensions $d>1$. Consider, for example, the random-trapping-model expression for the diffusion coefficient

$$D_0=\langle 1/w\rangle^{-1}\,, \tag{1.1}$$

where w is the hopping rate and angular brackets denote an ensemble average over the hopping rates. Equation (1.1) for D_0 is exact for the random-trapping model in any spatial dimension.[13,14] On the other hand, only for $d=1$ does (1.1) give the correct diffusion coefficient for the random-hopping model.[1,8,9] Perhaps the most striking difference between the two models lies in the fact that the random-hopping model is applicable to dynamical percolation, which arises whenever the distribution function contains a variable but finite concentration of zero jump rates.[6,7] As normally implemented, however, the random-trapping model can never lead to a percolation transition. Indeed, from (1.1) it is straightforward to show that any finite concentration of zero jump rates causes the diffusion constant to vanish. For $d>1$ the exact form of D_0 for the random-hopping model is a complex, and in fact, unknown function of the moments of w. Effective-medium theories have been developed for the random-hopping model,[6,7,10–12] and approximate expressions obtained for the diffusion coefficient, but theories which provide systematic corrections to the effective-medium results have proven elusive.

We demonstrate in this paper a new approach to the random-hopping model in two dimensions which does offer the possibility of yielding systematic corrections to transport properties as calculated in effective-medium theory. The method is based, in part, upon a new kind of generating function, which we adapt from an exact calculation performed recently by Kundu and Phillips[13] for the random-trapping model. Starting from an appropriate master equation we introduce *two* generating functions to describe the displacements along each direction of a square lattice. Analysis of the resultant equations leads to information about the long-time transport properties of the system. We defer to a future publication a full treatment of the general random-hopping model and, instead, consider in detail a system with fluctuating transfer rates in one direction and uniform rates in the other. For this system we investigate general effects arising from strong transport anisotropy, recovering exactly the limits in which transport in the uniform direction is infinitely fast $(D_0=\langle w\rangle)$ and infinitely slow $(D_0=\langle 1/w\rangle^{-1})$, the latter being the exact one-dimensional result. When we require that the long-time properties of the system be iso-

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tropic we obtain expressions which are, to lowest order in the fluctuating rates, identical in most cases to those obtained in effective-medium calculations, but which may, for this particular model, be systematically improved by considering successively higher-order terms in a general expansion. These results include the existence of a percolation-type transition for appropriate distributions. A simple modification to the theory is described which allows us to recover the exact effective-medium-theory results for the percolation problem.

In the next section we introduce the random master equation and the definitions of the generating functions that we will use to analyze the problem. These lead straightforwardly to coupled integral equations for the generating functions. We adapt these equations in Sec. III to treat the case where there are uniform rates in one direction and fluctuating rates in the other. An analysis of that problem follows. Emphasis is placed upon the long-time value of the diffusion constant for displacements in the fluctuating direction and on the form of low-frequency corrections. We conclude with a summary of the major results of the paper.

II. EQUATIONS FOR THE GENERATING FUNCTIONS

We now proceed to define and obtain a set of coupled integral equations for the two generating functions which form the basis of our calculation. We start with the master equation

$$\dot{p}_{n,m}=r_{n+1,m}(p_{n+1,m}-p_{n,m})-r_{n,m}(p_{n,m}-p_{n-1,m})+w_{n,m+1}(p_{n,m+1}-p_{n-m})-w_{n,m}(p_{n,m}-p_{n,m-1}) \quad (2.1)$$

for the probabilities $p_{n,m}(t)$ of finding the transport particle at the (n,m)th site of a two-dimensional square lattice at time t. In (2.1) the $r_{n,m}$ and $w_{n,m}$ are random nearest-neighbor rate constants governing hops in the x and y directions, respectively. They are presumed to be independent and distributed according to functions $\rho_x(r)$ and $\rho_y(w)$. Note that, by construction, the rate constant from a given site to its neighbor is equal to that for hops in the opposite direction, thus defining a random barrier, as opposed to a random-well problem.[5]

We now introduce two new quantities $P_{n,m}(t)\equiv p_{n,m}-p_{n-1,m}$ and $Q_{n,m}(t)\equiv p_{n,m}-p_{n,m-1}$ which permit us to write the mean-square displacement as[17,13]

$$\dot{x}=-\sum_{n,m} r_{n,m}P_{n,m} \ , \quad (2.2)$$

$$\dot{y}=-\sum_{n,m} w_{n,m}Q_{n,m} \ , \quad (2.3)$$

$$\dot{x}^2=-2\sum_{n,m} nr_{n,m}P_{n,m}+\sum_{n,m} r_{n,m}P_{n,m} \ , \quad (2.4)$$

$$\dot{y}^2=-2\sum_{n,m} mw_{n,m}Q_{n,m}+\sum_{n,m} w_{n,m}Q_{n,m} \ . \quad (2.5)$$

Here x and y are the (dimensionless) mean positions of the particle along the two orthogonal directions of the lattice, and the total mean-square displacement is $r^2=x^2+y^2$. (These definitions naturally refer to averages taken over walks with a fixed realization of the rates $r_{n,m}$ and $w_{n,m}$. We shall continue to denote averages taken over the ensemble of rates by angular brackets, such as $\langle r^2\rangle(t)$, etc.)

We now introduce two generating functions

$$g_1(\mathbf{k},t)=\sum_{n,m} r_{n,m}P_{n,m}\exp(ik_1n)\exp(ik_2m) \equiv \sum_{\mathbf{n}} r_{\mathbf{n}}P_{\mathbf{n}}e^{i\mathbf{k}\cdot\mathbf{n}} \ , \quad (2.6)$$

$$g_2(\mathbf{k},t)=\sum_{n} w_{\mathbf{n}}Q_{\mathbf{n}}e^{i\mathbf{k}\cdot\mathbf{n}} \quad (2.7)$$

for displacements along the x and y directions, wherein k_1 and k_2 are the x and y components of the reciprocal-lattice vector $\mathbf{k}$, and $\mathbf{n}$ denotes a direct-lattice vector with components n and m. With these definitions it is straightforward to show, for example, that

$$\dot{x}(t)=g_1(0,t)$$

and

$$\dot{x}^2(t)=\lim_{k\to 0}[g_1(\mathbf{k},t)+2i\partial g_1(\mathbf{k},t)/\partial k_1] \ . \quad (2.8)$$

Similar equations in terms of g_2 and its derivative with respect to k_2 hold for y and y^2. From Eq. (2.8) we see that the components of the diffusion constant, if they exist, may be readily obtained from a knowledge of g_1 and g_2. For example,

$$2D_{yy}=\lim_{t\to\infty}\lim_{\mathbf{k}\to 0}[g_2(\mathbf{k},t)+2i\partial g_2(\mathbf{k},t)/\partial k_2] \ . \quad (2.9)$$

We comment in passing that the generating functions introduced in (2.6) and (2.7)—which are precisely of the same form as those introduced by Kundu and Phillips[13] in their analysis of the random-trapping problem—differ from the generating functions typically used in lattice-diffusion problems of this sort by the inclusion of the factors $r_{n,m}$ or $w_{n,m}$ in the sum over lattice vectors. It is precisely this feature which in the random-trapping problem facilitates a straightforward expansion of the mean-square displacement in terms of the *inverse* moments of the hopping rates. In the present context we note from (2.8) that there is an additional simplification over standard treatments in the fact that only first derivatives of the generating function are required to obtain the mean-square displacement, rather than the usual two. As we shall see, this simplifies the extraction of long-time properties considerably inasmuch as the remaining derivative becomes trivial to perform.

From the definitions (2.6) and (2.7), the definitions of $P_{n,m}(t)$ and $Q_{n,m}(t)$, and the original master equation (2.1), it is a simple matter to obtain the collective equations of motion

$$\sum_{\mathbf{n}}\exp(i\mathbf{k}\cdot\mathbf{n})\dot{P}_{\mathbf{n}}=-2(1-\cos k_1)g_1(\mathbf{k},t)-[1-\exp(ik_1)][1-\exp(-ik_2)]g_2(\mathbf{k},t) \ ,$$
$$\sum_{\mathbf{n}}\exp(i\mathbf{k}\cdot\mathbf{n})\dot{Q}_{\mathbf{n}}=-2(1-\cos k_2)g_2(\mathbf{k},t)-[1-\exp(ik_2)][1-\exp(-ik_1)]g_1(\mathbf{k},t) \ , \quad (2.10)$$

which relate the time derivatives of $P_{\mathbf{n}}$ and $Q_{\mathbf{n}}$ to the two generating functions. We now wish to take Laplace transforms over the time variable. Consider first the term

$$\int_0^\infty dt\, e^{-\varepsilon t} \sum_{\mathbf{n}} \exp(i\mathbf{k}\cdot\mathbf{n})\dot{P}_{\mathbf{n}} = \varepsilon \sum_{\mathbf{n}} \exp(i\mathbf{k}\cdot\mathbf{n}) f_{\mathbf{n}}(\varepsilon) - F_1(\mathbf{k},0) \ , \quad (2.11)$$

where $f_{\mathbf{n}}(\varepsilon)$ is the Laplace transform of $P_{\mathbf{n}}(t)$ and the quantity

$$F_1(\mathbf{k},0) = \sum_{\mathbf{n}} \exp(i\mathbf{k}\cdot\mathbf{n}) P_{\mathbf{n}}(0) = [1-\exp(ik_1)] \quad (2.12)$$

arises from the initial conditions, $p_{n,m}(0)=\delta_{n,0}\delta_{m,0}$. The first term on the right-hand side of (2.11) can now be rewritten as

$$\varepsilon \sum_{\mathbf{n}} \exp(i\mathbf{k}\cdot\mathbf{n})[(1/r_{\mathbf{n}})-(1/c)+(1/c)]r_{\mathbf{n}} f_{\mathbf{n}}(\varepsilon) = sG_1(\mathbf{k},\varepsilon) + (2\pi)^{-2}\int d\mathbf{k}'\, sA_1(\mathbf{k},\mathbf{k}')G_1(\mathbf{k}',\varepsilon) \ , \quad (2.13)$$

where $G_1(\mathbf{k},\varepsilon)$ is the Laplace transform of the generating function $g_1(\mathbf{k},t)$, the kernel A_1 is defined by

$$A_1(\mathbf{k},\mathbf{k}') = (1/c)\sum_{\mathbf{n}} [(c/r_{\mathbf{n}})-1]\exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}] \ , \quad (2.14)$$

$s=\varepsilon/c$ is a scaled Laplace variable, and integrations over wave vectors are to be understood as running over the first Brillouin zone $-\pi \le k_1, k_2 \le \pi$. The quantity c is an as-of-yet unspecified constant which we have introduced into the equations, anticipating the possibility that at long times (or over large enough length scales) the system will behave as a uniform system with a frequency-independent diffusion constant.[9] Similar manipulations may be performed upon the term in (2.10) involving $Q_{\mathbf{n}}(t)$ which allow us to write the following coupled equations for the Laplace transforms, respectively, of the generating functions $g_1(\mathbf{k},t)$ and $g_2(\mathbf{k},t)$,

$$G_1(\mathbf{k},\varepsilon) = a_1(\mathbf{k},\varepsilon) - b_1(\mathbf{k},\varepsilon)G_2(\mathbf{k},\varepsilon) - \int d\mathbf{k}'\, \mathcal{K}(\mathbf{k},\mathbf{k}',\varepsilon)G_1(\mathbf{k}',\varepsilon) \ , \quad (2.15)$$

$$G_2(\mathbf{k},\varepsilon) = a_2(\mathbf{k},\varepsilon) - b_2(\mathbf{k},\varepsilon)G_1(\mathbf{k},\varepsilon) - \int d\mathbf{k}'\, \mathcal{J}(\mathbf{k},\mathbf{k}',\varepsilon)G_2(\mathbf{k}',\varepsilon) \ , \quad (2.16)$$

where

$$a_1 = [1-\exp(ik_1)]/[s+2(1-\cos k_1)] \ , \quad (2.17)$$

$$a_2 = [1-\exp(ik_2)]/[s+2(1-\cos k_2)] \ , \quad (2.18)$$

$$b_1 = [1-\exp(ik_1)][1-\exp(-ik_2)] \times [s+2(1-\cos k_1)]^{-1} \ , \quad (2.19)$$

$$b_2 = [1-\exp(ik_2)][1-\exp(-ik_1)] \times [s+2(1-\cos k_2)]^{-1} \ , \quad (2.20)$$

$$\mathcal{K}(\mathbf{k},\mathbf{k}',\varepsilon) = \frac{s(2\pi)^{-2}}{s+2(1-\cos k_1)} \sum_{\mathbf{n}} \alpha_{\mathbf{n}} \exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}] \ , \quad (2.21)$$

$$\mathcal{J}(\mathbf{k},\mathbf{k}',\varepsilon) = \frac{s(2\pi)^{-2}}{s+2(1-\cos k_2)} \sum_{\mathbf{n}} \beta_{\mathbf{n}} \exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}] \ , \quad (2.22)$$

and where $\alpha_{\mathbf{n}} \equiv (c/r_{\mathbf{n}})-1$; $\beta_{\mathbf{n}} \equiv (c/w_{\mathbf{n}})-1$.

It should be obvious that the solutions to the Eqs. (2.15) and (2.16) cannot depend explicitly upon the constant c, which is at this point somewhat arbitrary. However, by choosing c judiciously we can greatly affect the convergence properties of the equations that result from iteration of (2.15) and (2.16). It is to be emphasized, however, that Eqs. (2.15) and (2.16) are exact regardless of the value chosen for c.

III. UNIFORM RATES ALONG ONE AXIS

It is straightforward, starting from (2.15) and (2.16), to systematically decouple G_1 from G_2 and thereby using (2.8), investigate general features of the usual random-hopping model. We intend to present results based upon this approach in a separate publication. For the present, however, we wish to use (2.15) and (2.16) as the starting point to investigate a special case of the more general random-hopping model, a case which is certainly more tractable than the full problem, but one from which significant insights can still be obtained. We consider, specifically, the situation in which the rates associated with hops in the x direction are sharp, that is, take on a single value r, which we, at this point, set equal to the constant c introduced in the last section. This allows us to investigate how transport in the fluctuating direction is affected by hops perpendicular to that axis. For example, particles faced with a (locally) vanishing hopping rate in the y direction can "get around" the obstacle, by making hops in the x direction until a more favorable environment is found.

Under the assumptions stated above Eqs. (2.15) and (2.16) simplify, since the kernel $\mathcal{K}(\mathbf{k},\mathbf{k}',\varepsilon)$ now vanishes identically. As a result, we obtain

$$G_1 = a_1 - b_1 G_2 \ , \qquad G_2 = a_2 - b_2 G_1 - JG_2 \ , \quad (3.1)$$

in which we have suppressed the dependence upon $\mathbf{k}$ and ε, and where

$$J\{\cdots\} \equiv \int d\mathbf{k}'\, \mathcal{J}(\mathbf{k},\mathbf{k}',\varepsilon)\{\cdots\}_{\mathbf{k}'} \ .$$

The motion along the x direction, being uniform, is now strictly diffusive with diffusion constant c. Hence, insofar as we are primarily interested in displacements along the (fluctuating) y direction, we may formally solve (3.1) for the relevant generating function G_2,

$$G_2 = [1+(1-b_2b_1)^{-1}J]^{-1}(1-b_2b_1)^{-1}(a_2-b_2a_1) \ . \quad (3.2)$$

One could now proceed to analyze Eq. (3.2), which is an integral equation involving the operator J, by expanding the factor involving J on the right-hand side, and performing the averages term by term using $\rho_y(w)$. Instead, we have decided to follow a slightly different approach by separating G_2 into two parts: the first part is independent of ε upon averaging and describes the very-long-time properties of the system. It defines the diffusion constant (when it exists) for displacements along the y direction. The second term, which does depend upon ε, describes the approach to diffusion (it contains what are sometimes referred to, perhaps misleadingly, as "non-Markovian" corrections). The details of this separation, which is somewhat lengthy albeit straightforward, are provided in Appendix A.

The integral equation which results may be expressed in the form $G=[\mathscr{G}^0+\mathscr{G}^1(\varepsilon)]f$, where the operators $\mathscr{G}^0$ and $\mathscr{G}^1(\varepsilon)$ make manifest the aforementioned separation, and where

$$f(\mathbf{k},\varepsilon)=\frac{1-\exp(ik_2)}{(s/2)\langle 1-a\rangle+1-\langle 1-a\rangle\cos k_1-\langle a\rangle\cos k_2}\ , \tag{3.3}$$

is a function that is independent of the fluctuations. In (3.3) we have introduced the quantity $\langle a\rangle=\langle a_{\mathbf{n}}\rangle \equiv\langle w_{\mathbf{n}}/(c+w_{\mathbf{n}})\rangle$.

We note that in the zero-wave-vector limit, the quantity f goes to zero and its derivative with respect to k_2 becomes independent of ε. This, coupled with the fact that the averaged system is translationally invariant (and hence that $\langle\mathscr{G}^0\rangle$ (as well as $\langle\mathscr{G}^1\rangle$) is a diagonal operator [i.e., $\langle\mathscr{G}^0\rangle_{\mathbf{k},\mathbf{k}'}=\mathscr{G}^0(\mathbf{k})\delta(\mathbf{k}-\mathbf{k}')$]), means that we can use (2.9) to express the zero-frequency diffusion constant for the y direction in a particularly simple form:

$$\begin{aligned} D_{yy}&=\lim_{\mathbf{k}\to 0}\lim_{\varepsilon\to 0}[i\mathscr{G}^0(\mathbf{k})\partial\varepsilon f(\mathbf{k},\varepsilon)/\partial k_2]\\ &=2\langle 1/(c+w)\rangle^{-1}\mathscr{G}^0(0)\ . \end{aligned} \tag{3.4}$$

Similarly, the *approach* to diffusion can be expressed solely in terms of the small-ε behavior in the zero-wave-vector limit of $\mathscr{G}^1(\mathbf{k}\varepsilon)$, by which we denote the diagonal part of the averaged operator $\langle\mathscr{G}^1\rangle$. The operators $\mathscr{G}^0$ and $\mathscr{G}^1$ are given explicitly by (see Appendix A for a derivation):

$$2\mathscr{G}^0=B+\langle 1-a\rangle^{-1}\delta B\Delta_1\delta B(1-\Delta_1\delta B)^{-1}+\Xi_0\ , \tag{3.5}$$

$$2\langle 1-a\rangle\mathscr{G}^1=\delta B(1-\Delta_1\delta B)^{-1}\Gamma(1-\Gamma)^{-1}+\Xi_1\ , \tag{3.6}$$

where $\Delta_1=\Delta(s=0)$, $\Delta_2=\Delta-\Delta_1$, $\Gamma=\Delta_2\delta B(1-\Delta_1\delta B)^{-1}$,

$$[B]_{\mathbf{k},\mathbf{k}'}=(2\pi)^{-2}\sum_{\mathbf{n}}a_{\mathbf{n}}\exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}]\ , \tag{3.7}$$

$$[\delta B]_{\mathbf{k},\mathbf{k}'}=(2\pi)^{-2}\sum_{\mathbf{n}}\delta a_{\mathbf{n}}\exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}]\ , \tag{3.8}$$

and

$$\Delta(s,\mathbf{k})=\frac{s/2+\cos k_2-\cos k_1}{(s/2)\langle 1-a\rangle+1-\langle a\rangle\cos k_2-\langle 1-a\rangle\cos k_1}\ . \tag{3.9}$$

The quantity $\delta a_{\mathbf{n}}$ appearing in (3.8) is defined as $a_{\mathbf{n}}-\langle a\rangle$, and the operators Ξ_0 and Ξ_1 have the following definitions:

$$\Xi_0=\langle a\rangle\langle 1-a\rangle^{-1}\delta B\ ,$$

$$\Xi_1=\langle a\rangle\langle 1-a\rangle(\Delta-\Delta_{\mathbf{k}=0})\delta B(1-\Delta\delta B)^{-1}\ .$$

After averaging over the fluctuations the matrix δB, and hence Ξ_0, is zero. Also, in taking the limit $\mathbf{k}\to 0$, the factor involving $\Delta-\Delta_{\mathbf{k}=0}$ causes Ξ_1 to vanish as well. Neither of these terms can contribute, therefore, to either the diffusion constant or the frequency corrections, and so we ignore both of these terms in the discussion which follows.

The operators $(1-\Delta_1\delta B)^{-1}$ and $(1-\Gamma)^{-1}$ appearing in (3.5) and (3.6) may now be expanded in a geometric series and averaged over the fluctuating rates in the usual fashion to obtain for $D_{yy}(0)$ and its frequency-dependent corrections a systematic expansion in moments of the deviations $(\delta a_{\mathbf{n}})$ of the spatially fluctuating quantity $a_{\mathbf{n}}=w_{\mathbf{n}}/(c+w_{\mathbf{n}})$ from its average value. It will be noted that $(\delta a_{\mathbf{n}})$ is a reasonable expansion parameter in this regard insofar as the inequality $|\delta a_{\mathbf{n}}|\le 1$ is always satisfied.

A. The diffusion constant $D_{yy}(0)$

Let us first consider the expansion (3.5) for the zero-frequency, i.e., $\varepsilon\to 0$, limit of the generating function, from which we may obtain the diffusion constant D_{yy} through Eq. (3.4). By expanding (3.5) we obtain

$$\begin{aligned} 2\langle\mathscr{G}^0\rangle=\langle a\rangle&+\langle 1-a\rangle^{-1}\langle\delta B\Delta_1\delta B\rangle\\ &+\langle 1-a\rangle^{-1}\langle\delta B\Delta_1\delta B\Delta_1\delta B\rangle\\ &+\langle 1-a\rangle^{-1}\langle\delta B\Delta_1\delta B\Delta_1\delta B\Delta_1\delta B\rangle+\cdots\ . \end{aligned} \tag{3.10}$$

While it is possible, in principle, to continue this expansion to any desired order in the fluctuations, let us consider first the lowest-order theory that results when we truncate (3.10) following the first term and write $2\langle\mathscr{G}^0\rangle=\langle a\rangle$. This approximation to the full sum (3.10) yields the following expression for D_{yy}:

$$\begin{aligned} D_{yy}&=\langle a\rangle\langle 1/(c+w)\rangle^{-1}\\ &=\langle w/(c+w)\rangle\langle 1/(c+w)\rangle^{-1}\ . \end{aligned} \tag{3.11}$$

Equation (3.11) demonstrates explicitly how the diffusion constant for displacements in the y direction depends parametrically upon the uniform value c of the hopping rate in the x direction. By changing the value of this rate, therefore, it becomes possible to investigate effects of transport anisotropy which might arise in amorphous systems.

As an example of obvious interest, let us consider the limit $c\to 0$, in which transport in the x direction stops and the system reduces to a set of isolated random one-dimensional chains. In this limit $a_{\mathbf{n}}\equiv 1$ for all $\mathbf{n}$, and the fluctuations $(\delta a_{\mathbf{n}})$ vanish identically. The zeroth-order result then becomes *exact* and we recover the known one-dimensional result

$$D_{yy}=\langle 1/w\rangle^{-1},\ \ d=1\ , \tag{3.12}$$

which was obtained previously for both random-trapping[13] and random-hopping models.[8,9] We point out, however, that in the present case the result follows quite naturally from a systematic expansion of the generating function and not as a consequence of a self-consistent approximation. [Indeed, the result could have been obtained in an even more straightforward way by setting $b_2=0$ in the original coupled equations (3.1) and proceeding along similar lines.]

As a second interesting example, consider the opposite limit in which the motion along the x direction is extremely rapid, i.e., $c\to\infty$. In this limit $a_n\to 0$, hence the fluctuations δa_n also vanish and presumably, again, we find the exact result

$$D_{yy} \underset{c\to\infty}{\longrightarrow} \langle w \rangle \,. \tag{3.13}$$

The result (3.13) can be interpreted physically as a kind of motional averaging produced by the rapid motion along the x direction, which forces the moving particle to experience the "average" environment for hops in the y direction.

In between these two limits of course there is a wide range of behavior, and for a given distribution $\rho_y(w)$ we expect that there will be a unique value of c which will make the two-dimensional system *isotropic* at long times, that is, which makes $\langle x^2(t)\rangle \sim \langle y^2(t)\rangle$ at $t\to\infty$. Since displacements in the x direction are uniform in time, this state of eventual isotropy is determined by equating the respective long-time diffusion constants in each direction:

$$c=(\langle a\rangle+\langle 1-a\rangle^{-1}\langle\delta B\Delta_1\delta B\rangle + \langle 1-a\rangle^{-1}\langle\delta B\Delta_1\delta B\Delta_1\delta B\rangle+\cdots)\langle 1/(c+w)\rangle^{-1}\,. \tag{3.14}$$

If we again truncate the series at lowest order we obtain the approximate isotropy condition:

$$\langle c/(c+w)\rangle=\langle a\rangle=\langle w/(c+w)\rangle = \tfrac{1}{2}\,. \tag{3.15}$$

This, it may be recognized, is precisely the condition which arises in the *effective-medium approximation* (EMA) for determining the diffusion constant for a system which has fluctuating rates in *both* directions.[18] To lowest order in our calculation, then, we recover for this system the numerous results that have been obtained within the EMA for many different classes of distribution functions. In particular, we note that for a percolation distribution $\rho_y(w)=p\delta(w-W)+(1-p)\delta(w)$, where $1-p$ is the fraction of broken bonds and W is the rate associated with a normal bond, the condition of long-time transport isotropy *induces* a percolation-type transition on this system with a percolation concentration $p_c=\frac{1}{2}$, the exact result for two dimensions. Indeed, at the value $p_c=\frac{1}{2}$, the diffusion constant for the y direction D_{yy} vanishes to all orders in the fluctuations. For $p>p_c$ the diffusion constant predicted by (3.15) takes the form

$$D(p)=c=2W(p-p_c),\quad p\ge p_c\,. \tag{3.16}$$

which is well known from EMA.[6]

We have obtained Eqs. (3.15) and (3.16) from the lowest-order term in the expansion of the generating function. It is also possible to consider corrections to (3.16) which arise from the higher-order terms. We emphasize that although our results are similar to those of EMA, the corrections to (3.16) apply only to the model system we have been considering (with uniform motion along one direction) and do *not* necessarily converge to the result for the fully fluctuating lattice. However, they are useful as a rough indication of the rate of convergence of our expansion and as an approximate analytic measure of the magnitude of the corrections that arise in the fully fluctuating case.

We consider first the degree of anisotropy Θ that actually arises in this system when we impose the approximate isotropy condition (3.15). The next nonzero correction is of order $\langle\delta^2 a\rangle\langle\delta^3 a\rangle$ and we find

$$\Theta\equiv(c-D_{yy})/c=-4\xi\langle\delta^3 a\rangle\langle\delta^2 a\rangle+O(\langle\delta^7 a\rangle) \tag{3.17}$$

$$=\xi(1/8p^3)(1-p)^2(2p-1)\,, \tag{3.18}$$

the latter being the percolative result. The constant ξ appearing in (3.17) and (3.18) arises from the averages of the matrices appearing in (3.14), and is defined explicitly in Appendix B. A numerical evaluation of the lattice sums gives $\xi\approx 0.36$. The results suggest that (3.15) tends to slightly *overestimate* the diffusion constant but is still valid to within 1%.

An alternate approach to the correction terms is obtained by simply truncating the full isotropy condition defined by (3.14) at some high-order fluctuation correction. The isotropy condition correct to fifth order linearized around $\langle a\rangle=\frac{1}{2}$,

$$c=[\langle a\rangle+\langle 1-a\rangle^{-1}(\gamma\langle\delta^2 a\rangle\langle 2a-1\rangle+\beta\langle\delta^2 a\rangle^2\langle 2a-1\rangle+\xi\langle\delta^2 a\rangle\langle\delta^3 a\rangle)]\langle 1/(c+w)\rangle^{-1}\,, \tag{3.19}$$

contains two new constants $\gamma\approx 0.73$ and $\beta\approx 1.1$ which are defined in Appendix B. For the percolative case (3.19) yields a fifth-order equation for c which may be solved numerically. The results of this procedure again agree with (3.15) to within a few percent and vanish at $p_c=\frac{1}{2}$.

B. Frequency-dependent corrections: The approach to diffusion

We now focus on the approach of the transport properties to the long-time diffusive limit. This information is contained in the expansion of Eq. (3.6) for the operator $\mathscr{G}^1$, and we therefore define a frequency-dependent diffusion constant through the relation

$$D_{yy}(\varepsilon)-D_{yy}(0)=(\varepsilon^2/2)\langle y^2(\varepsilon)\rangle$$
$$=2\mathscr{G}^1(\varepsilon,\mathbf{k}=0)\langle 1/(c+w)\rangle^{-1} . \quad (3.20)$$

In light of our discussion regarding the diffusion constant, it should be clear that the manner in which the system approaches the diffusive limit at long times will depend strongly upon the value of the uniform hopping rate c. Let us consider some of the examples discussed in the preceding section. For the case in which the motion along the uniform x direction is the fastest ($c\to\infty$) the matrix $\delta B\to 0$. It follows, therefore, from (3.6) that in this limit the frequency-dependent corrections vanish entirely, and the system is strictly diffusive in the y direction.

Obtaining the proper frequency corrections for the one-dimensional limit,[8,9] in which $c\to 0$, is slightly more complicated but still straightforward. It is relatively easy to show that in this limit the operator $\Delta_1\delta B$ goes to zero as c itself. Moreover, expanding the expression (3.8) for small c allows us to write $\delta B(\mathbf{k},\mathbf{k}')\sim -\langle c/w\rangle\delta b(\mathbf{k},\mathbf{k}')$ where

$$\delta b(\mathbf{k},\mathbf{k}')=(2\pi)^{-2}\sum_{\mathbf{n}}(w_{\mathbf{n}}^{-1}/\langle w^{-1}\rangle-1)\exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}] . \quad (3.21)$$

We now restrict our attention to those distributions $\rho_y(w)$ for which the inverse moments of w exist. Thus as $c\to 0$, we can use (3.21) and the fact that in this limit $\langle 1-a\rangle\sim\langle c/w\rangle$ to write (3.6) in the asymptotic form

$$2\mathscr{G}^1=\langle c/w\rangle\delta b\Delta\delta b(1-\langle c/w\rangle\Delta\delta b)^{-1} . \quad (3.22)$$

To obtain (3.22) we used the fact that $\Delta_1\delta B\to 0$ for small c. Hence $\Delta_2\delta B=(\Delta-\Delta_1)\delta B=\Delta\delta B$ in the small-c limit. From the definition (3.9) of Δ, we find that

$$\lim_{c\to 0}\langle c/w\rangle\Delta(\mathbf{k},\varepsilon)=\sigma[\sigma+2(1-\cos k_2)]^{-1}$$
$$\equiv\Delta'(\mathbf{k},\varepsilon) , \quad (3.23)$$

where $\sigma=\langle\varepsilon/w\rangle$. Thus, (3.22) becomes

$$2\mathscr{G}^1=\delta b\Delta'\delta b(1-\Delta'\delta b)^{-1}$$
$$=\delta b\Delta'\delta b+\delta b\Delta'\delta b\Delta'\delta b+\cdots . \quad (3.24)$$

Now each factor of Δ' brings with it an additional multiplicative factor of σ (i.e., of ε), so that in the small frequency limit the leading contribution comes from the first term in the expansion (3.24). Hence, we obtain for small ε

$$D_{yy}(\varepsilon)-D_{yy}(0)\sim\langle 1/w\rangle\langle\delta^2(1/w)\rangle(2\pi)^{-2}$$
$$\times\int d\mathbf{k}\{\sigma/[\sigma+2(1-\cos k_2)]\}$$
$$\sim\tfrac{1}{2}\langle 1/w\rangle^{-5/2}\langle\delta^2(w^{-1})\rangle\varepsilon^{1/2}+O(\varepsilon) , \quad (3.25)$$

which has also been obtained by other methods for both the random-trapping[13] and random-hopping[8,9] models in one dimension.

Finally, let us turn to the isotropic limit which we will again treat to low orders in the fluctuations of $\delta a_{\mathbf{n}}$. If we expand (3.6) in terms of the operator Γ and substitute into (3.20), we obtain

$$D_{yy}(\varepsilon)-D_{yy}(0)=c\langle\delta B(1-\Delta_1\delta B)^{-1}(\Gamma+\Gamma^2+\Gamma^3+\cdots)\rangle\langle(1-a)\rangle^{-2} . \quad (3.26)$$

From the definition following (3.6) it will be seen that the operator Γ is proportional to the function Δ_2. This function has been defined previously to be

$$\Delta_2(\varepsilon,\mathbf{k})=\Delta-\Delta_1=\frac{(s/2)(1-\cos k_2)}{[(s/2)\langle 1-a\rangle+1-\langle a\rangle\cos k_2-\langle 1-a\rangle\cos k_1](1-\langle a\rangle\cos k_2-\langle 1-a\rangle\cos k_1)} . \quad (3.27)$$

The proportionality of Δ_2 (and therefore of Γ) to s implies that the terms in (3.26) containing successively higher powers of Γ will go to zero as successively higher-order functions of ε. The leading order correction to the zero-frequency limit, therefore, comes from the first term in (3.26) which contains only one factor of Γ,

$$D_{yy}(\varepsilon)-D_{yy}(0)\sim c\langle\delta B[1-\Delta_1\delta B]^{-1}\Delta_2\delta B[1-\Delta_1\delta B]^{-1}\rangle$$
$$\times\langle 1-a\rangle^{-2} . \quad (3.28)$$

To proceed, we now expand each factor of $[1-\Delta_1\delta B]^{-1}$, and collect terms of the same order in moments of $\langle\delta^n a\rangle$. The first two terms are $c\langle\delta B\Delta_2\delta B\rangle\langle 1-a\rangle^{-2}$ which is of order $\langle\delta^2 a\rangle$ and $c\langle\delta B\Delta_1\delta B\Delta_2\delta B+\delta B\Delta_2\delta B\Delta_2\delta B\rangle\langle 1-a\rangle^{-2}$ which appears to third order in the fluctuations. Let us now consider the expressions that result when we define c by the lowest-order isotropy condition (3.15), for which $\langle a\rangle=\frac{1}{2}$. This condition makes all terms of odd order in the fluctuations vanish as a consequence of a resultant symmetry in the integrals (see Appendix B). Retaining only the second-order term from above then yields the approximation

$$D_{yy}(\varepsilon)-D_{yy}(0)\sim c\langle\delta^2 a\rangle\langle 1-a\rangle^{-2}(2\pi)^{-2}$$
$$\times\int\Delta_2(\mathbf{k},\varepsilon)d\mathbf{k} . \quad (3.29)$$

Substituting the definition of $\Delta_2(\mathbf{k},\varepsilon)$ into (3.29) and performing the integral, we obtain

$$D_{yy}(\varepsilon)-D_{yy}(0)\sim\frac{\langle\delta^2 a\rangle 4\varepsilon\mathbf{K}(4/(s+4))}{\pi(s+4)\langle 1-a\rangle^2} , \quad (3.30)$$

where $\mathbf{K}(x)$ is the complete elliptic integral of the first kind. Using the known asymptotic properties of the elliptic integral, we finally obtain

$$D_{yy}(\varepsilon)-D_{yy}(0)\sim-(2/\pi)\langle\delta^2 a\rangle\varepsilon\ln\varepsilon+O(\varepsilon)\ , \quad (3.31)$$

which exhibits the $\varepsilon\ln\varepsilon$ dependence of the leading frequency correction characteristic feature of hopping transport in two-dimensional disordered systems.[6,13,19]

In deriving (3.31) we have assumed that the quantity c defining the motion in the uniform direction was a constant, that is, independent of ε. It comes as no surprise therefore that for specific distributions the coefficients of the frequency correction predicted by (3.31) *with a constant value of* c do not agree with the EMA results for the fully fluctuating system. For example, in the percolative case (3.31) yields $-2\langle\delta^2 a\rangle/\pi=-(1-p)/2\pi p$ for the coefficient of $\varepsilon\ln\varepsilon$, whereas the corresponding effective-medium result is $-[(1-p)/4\pi(p-p_c)]$. This disagreement stems from an actual difference between the physical models. To illustrate, as p approaches p_c, and $c\rightarrow 0$, the model treated here reduces to a set of isolated random chains consisting of clusters of connected sites separated by barriers. Absolutely no transport between the chains is possible. In an actual percolative system, however, even though the diffusion constant in the x direction goes to zero as $p\rightarrow p_c$, transport in this direction is still possible over length scales comparable to the mean interbarrier spacing. This residual transport will obviously influence the motion in the y direction in ways which we have already discussed.

The EMA approach of Webman[11] and Odagaki, Lax, and Puri[6] treats the uniform hopping rate as a frequency-dependent parameter, $c(\varepsilon)$. Because none of our derivations hinge on c being independent of frequency, we now treat c in the manner suggested by EMA theories.[6,11] To lowest order, the frequency-dependent diffusion coefficient in the y direction is

$$D_{yy}(\varepsilon)=c\langle a\rangle\langle 1-a\rangle^{-1}-(2\pi)^{-1}\langle 1-a\rangle^{-2}\langle\delta^2 a\rangle \times\varepsilon\ln\varepsilon+O(\varepsilon)\ . \quad (3.32)$$

Let us assume that $c(\varepsilon)=c_0+c_1(\varepsilon)$. The new isotropy condition,

$$[c_0+c_1(\varepsilon)]\langle 1-2a\rangle=-\langle 1-a\rangle^{-1}(2\pi)^{-1}\langle\delta^2 a\rangle\varepsilon\ln\varepsilon\ , \quad (3.33)$$

now requires that we equate diffusion constant as well as the approach to the diffusive limit along the two axes of the crystal. Substituting in the moments associated with the percolation distribution and retaining the lowest nonzero orders in ε and $c_1(\varepsilon)$, we find

$$D_{yy}(\varepsilon)=c_0+c_1(\varepsilon) =2(p-p_c)W-[(1-p)/4\pi(p-p_c)]\varepsilon\ln\varepsilon\ . \quad (3.34)$$

Thus, as in the EMA, to obtain sensible results for the fully fluctuating system it is necessary to assume a frequency-dependent rate.[6,11] The novel feature of the present derivation, however, is that we obtain the results by equating the long-time transport properties along different directions of the crystal, rather than as a result of averaging over *local* deviations from the effective medium.

IV. CONCLUSIONS

In this paper we have introduced a method for analyzing the transport properties of random-hopping models that is substantially different from most existing methods currently employed in the analysis of disordered systems. The method is innovative in two particular respects. First, it extends to random-hopping models a new type of generating function recently introduced by Kundu and Phillips for the random-trapping problem.[13] The utility of this approach is derived from a natural extension of expressions introduced by van Kampen[17] for the mean-square displacement of linear chains. We believe that generating functions of this kind can offer several advantages over transitional ones, especially in the treatment of strongly disordered systems.[13,20]

The second new feature of our method is the introduction of separate generating functions to describe displacements along each direction of the lattice. While this may at first sight appear to be somewhat of a complication, we believe that the process of decoupling the displacements in one direction from those in the other provides insight into important mechanisms that can affect hopping transport in amorphous systems. Indeed, this concept in part provides the motivation of our calculations of Sec. III, where we have specialized our method to treat the case in which transport along one of the directions is uniform. We find that in many respects, such as system resembles one with fluctuating rates in both directions, as is evidenced by our derivation of effective-medium results for the diffusion constant and characteristic low-frequency behavior.

It is of obvious interest to ask how well such a model actually represents a system with fluctuations in both directions, and in particular, how relevant the corrections we have calculated for the diffusion constant are to real percolating lattices and other disordered systems? Answering such questions obviously requires an analysis of the fully coupled equations (2.10) and (2.11) to which Eqs. (3.1) may be considered as an approximation. Indeed, one could take the view that our isotropic calculations of Sec. III represent an attempt to determine an effective medium for displacements along the x direction as seen by the fluctuating y direction. Seen in this light, our derivation of effective-medium-type expressions is not surprising, and in fact yields further insight into the meaning and applicability of effective-medium theory. Nonetheless, it seems certain that in simplifying (2.10) and (2.11) to the form of (3.1), we have neglected corrections that could prove important when the rates in the x direction are also allowed to fluctuate. We comment in closing that tentative calculations on the three-dimensional analog to this problem have yielded some interesting preliminary results. As in the two-dimensional case, the lowest-order results again recover the predictions of effective-medium theory—including the existence of a percolation threshold at the (incorrect) effective-medium value $p=\frac{1}{3}$. The correction terms which arise, however, unlike the situation we have presented here for the square lattice, do *not* vanish at the effective-medium value for p_c.

This suggests the possibility of obtaining a higher-order self-consistent (i.e., isotropic) condition of the type discussed in Sec. III which comes closer to predicting the actual value $p_c \approx \frac{1}{4}$.

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APPENDIX A: SEPARATION OF G_2

In this appendix we give the details regarding the separation of the generating function $G_2(\mathbf{k},\varepsilon)$, as it appears in (3.2), into the two components $\mathscr{G}^0 f$ and $\mathscr{G}^1 f$ given in (3.5) and (3.6). The process is complicated by the fact that many of the functions and matrices we are dealing with have zeros at $\mathbf{k}=0$, and thus do not have well-defined inverses in the zero-frequency limit. We start out by defining the function

$$a_3 = 2[(1-b_2 b_1)^{-1}(a_2 - b_2 a_1)] \ .$$

Given the definitions (2.17)—(2.20) we find

$$a_3(\mathbf{k}) = [1-\exp(ik_2)][(s/2)+2-\cos k_1 - \cos k_2]^{-1} \ . \tag{A1}$$

When substituted into (3.3) this yields $2G_2 = [1 + (1-b_2 b_1)^{-1} J]^{-1} a_3$. We are therefore led to consider the operator $M = 1 + (1-b_2 b_1)^{-1} J$. Substituting in from the definitions (2.17)—(2.20), we obtain

$$2[M]_{\mathbf{k},\mathbf{k}'} = (2\pi)^{-2} \sum_{\mathbf{n}} [(c/w_{\mathbf{n}})+1]\exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}] + \frac{(s/2)+\cos k_2 - \cos k_1}{(s/2)+2-\cos k_1 - \cos k_2}(2\pi)^{-2} \times \sum_{\mathbf{n}} \beta_{\mathbf{n}} \exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}] \ . \tag{A2}$$

Let us write this as $M = \frac{1}{2}[J_1 + J_2(s)]$, which implicitly defines J_1 and $J_2(s)$ through Eq. (A2). With this notation the generating function can then be written

$$G_2 = [J_1 + J_2(s)]^{-1} a_3 = J_1^{-1}(1 + J_2 J_1^{-1})^{-1} a_3 \ , \tag{A3}$$

where the inverse of J_1, which we denote by B, is well defined and given by Eq. (3.7). We now introduce an operator δB, and two functions L and Δ_0, which will allow us to express (A3) in a more convenient form. The operator δB, whose components are given explicitly in Eq. (3.8), represents the deviation of the operator B from its average value $\langle B \rangle = \langle a \rangle$; the functions L and Δ_0 are defined as follows:

$$L(\mathbf{k}) = \frac{(s/2)\langle 1-a \rangle + 1 - \langle a \rangle \cos k_2 - \langle 1-a \rangle \cos k_1}{(s/2)+2-\cos k_2 - \cos k_1} \ , \tag{A4}$$

$$\Delta_0(\mathbf{k}) = \frac{(s/2)+\cos k_2 - \cos k_1}{(s/2)+2-\cos k_2 - \cos k_1} \ . \tag{A5}$$

With these definitions, it is a straightforward exercise in algebra to express the operator $1 + J_2 J_1^{-1}$ appearing in (A3) in the form $L - \Delta_0 \delta B$. This leads to the following expression for the generating function:

$$2G_2 = B(L - \Delta_0 \delta B)^{-1} a_3 = B(1 - \Delta \delta B)^{-1} f \ , \tag{A6}$$

where the functions $\Delta = L^{-1}\Delta_0$ and $f = L^{-1} a_3$ are given explicitly by (3.9) and (3.3), respectively. Now, using the identity $(1-x)^{-1} = 1 + x(1-x)^{-1}$, with $x = \Delta\delta B$, along with the previously mentioned identity $B = \langle a \rangle + \delta B$, we obtain from (A6)

$$2G_2 = [B + B\Delta\delta B(1-\Delta\delta B)^{-1}]f = [B + \langle a \rangle \Delta\delta B(1-\Delta\delta B)^{-1} + \delta B\Delta\delta B(1-\Delta\delta B)^{-1}]f \ . \tag{A7}$$

When the second term in (A7) is averaged and the limit $k \to 0$ taken, the quantity Δ which multiplies the left of that expression will appear only in its $\mathbf{k}=0$ value $\langle 1-a \rangle^{-1}$. We therefore separate out this term and denote the deviation from it by Ξ_1:

$$2G_2 = [B + \langle a \rangle \langle 1-a \rangle^{-1} \delta B(1-\Delta\delta B)^{-1} + \delta B \Delta \delta B (1-\Delta\delta B)^{-1} + \Xi_1] f = [B + \langle a \rangle \langle 1-a \rangle^{-1} \delta B + \langle 1-a \rangle^{-1} \delta B \Delta \delta B (1-\Delta\delta B)^{-1} + \Xi_1] f \ , \tag{A8}$$

where the second line again follows from an application of the identity $(1-x)^{-1} = 1 + x(1-x)^{-1}$. We are now in a position to let $s \to 0$ in the remaining terms, thereby isolating the long-time behavior. This is most easily done by separating $\Delta(s,\mathbf{k})$ into components $\Delta_1(\mathbf{k}) = \Delta(s=0,\mathbf{k})$ and $\Delta_2 = \Delta - \Delta_1$. After only a slight amount of algebra we then obtain

$$2G_2 = (\mathscr{G}^0 + \mathscr{G}^1) f \ , \tag{A9}$$

where the operators $\mathscr{G}^0$ and $\mathscr{G}^1$ are defined explicitly in Eqs. (3.5)—(3.9).

APPENDIX B: CALCULATION OF AVERAGES

In this appendix we discuss the calculation of averages which appear in expansions such as (3.10), (3.14), and (3.24). For example, all terms required in the expansion (3.10) for the diffusion constant can be expressed in the general form

$$X_r = \langle \delta B (\Delta_1 \delta B)^{r-1} \rangle \ , \tag{B1}$$

which is of order r in the random matrix δB. After averaging, (B1) is a diagonal matrix in the $\mathbf{k}$ representation. This is just a statement of the translational invariance of the averaged system. Nonetheless, it is sometimes more convenient to work in real space. For example, the (n,m)th element of X_r is given by

$$[X_r]_{n,m}=\sum_{n_1,n_2,\ldots,n_{r-2}}\langle \delta a_n z_{n-n_1}\delta a_{n_1} z_{n_1-n_2}\delta a_{n_2}\cdots z_{n_{r-2}-m}\delta a_m\rangle$$
$$=\sum_{n_1,\ldots,n_{r-2}}\langle \delta a_n \delta a_{n_1}\cdots\delta a_m\rangle z_{n-n_1}z_{n_1-n_2}\cdots z_{n_{r-2}-m}\ , \tag{B2}$$

where $\delta a_n \delta_{n,m}$ is the $(\mathbf{n},\mathbf{m})$th element of δB and z_n is the discrete Fourier inverse of the function $\Delta_1(\mathbf{k})$. From the definition of Δ_1 as the $s\to 0$ limit of the function $\Delta(s,\mathbf{k})$ we find

$$z_n=\int_0^\infty \exp(-t)[I_{n_1}(\langle at\rangle)I'_{n_2}(\langle bt\rangle)-I'_{n_1}(\langle at\rangle)I_{n_2}(\langle bt\rangle)]dt\ , \tag{B3}$$

where $I_n(x)$ is the modified Bessel function, $\langle b\rangle=\langle 1-a\rangle$, $\langle a\rangle=\langle w/(c+w)\rangle$, and $I'_n(x)=\frac{1}{2}[I_{n+1}(x)+I_{n-1}(x)]$. We can now proceed to perform the averages in (B2) using the facts that $\langle \delta a_n\rangle=0$ and that the fluctuations on different sites are uncorrelated. Thus, e.g., $\langle \delta a_n \delta a_m\rangle=\langle \delta^2 a\rangle\delta_{n,m}$, etc. Let us consider some examples. The next three terms in the expansion of the diffusion constant beyond the first are

$$\langle \delta B\Delta_1\delta B\rangle_{n,m}=\langle \delta a_n\delta a_m\rangle z_0=\langle\delta^2 a\rangle z_0\delta_{n,m}\ , \tag{B4}$$

$$\langle \delta B\Delta_1\delta B\Delta_1\delta B\rangle_{n,m}=\sum_{n_1}\langle \delta a_n\delta a_{n_1}\delta a_m\rangle z_{n-n_1}z_{n_1-m}=\langle\delta^3 a\rangle (z_0)^2\delta_{n,m}\ , \tag{B5}$$

$$\langle \delta B\Delta_1\delta B\Delta_1\delta B\Delta_1\delta B\rangle_{n,m}=\sum_{n_1,n_2}\langle \delta a_n\delta a_{n_1}\delta a_{n_2}\delta a_m\rangle z_{n-n_1}z_{n_1-n_2}z_{n_2-m}$$
$$=\langle\delta^4 a\rangle(z_0)^3\delta_{n,m}+\langle\delta^2 a\rangle^2(z_0)^2 z_{n-m}+\langle\delta^2 a\rangle^2 z_0\left[\sum_{n_1}(z_{n_1})^2\right]\delta_{n,m}+\langle\delta^2 a\rangle^2(z_{n-m})^3\ . \tag{B6}$$

Notice that these are all functions of $|\mathbf{n}-\mathbf{m}|$ only, as one would expect of operators which are diagonal in $\mathbf{k}$ space. Let us now consider what happens to these terms when the approximate isotropy condition (3.15) is imposed. With this choice for the value c, the quantities $\langle a\rangle$ and $\langle b\rangle$ both become equal to $\frac{1}{2}$. Equation (B3) shows, however, that in this limit z_n becomes antisymmetric under interchange of the components n_1 and n_2. Therefore, when $\langle a\rangle=\langle b\rangle=\frac{1}{2}$ the quantity z_0, which appears throughout Eqs. (B4)–(B6), vanishes identically. In addition, when evaluating the diffusion constant we require only the $\mathbf{k}\to 0$ limit of the operators which appear in these equations. Thus terms such as the last one in (B6) which contain factors of z_{n-m} will end up being summed over $\mathbf{n}-\mathbf{m}$. The antisymmetry of z_n under interchange of components then insures that those terms which contain odd powers of z_{n-m} will vanish along with z_0. Indeed, it is possible in this way to show that *all* terms which are of even order in the fluctuations will vanish in the limit $\langle a\rangle=\langle b\rangle=\frac{1}{2}$.

In fact, the next nonzero contribution in this limit is of fifth order in the fluctuations and has the form

$$\langle\delta^2 a\rangle\langle\delta^3 a\rangle\left[\sum_{n_1}(z_{n_1})^4\right]\delta_{n,m}=\langle\delta^2 a\rangle\langle\delta^3 a\rangle\xi\ . \tag{B7}$$

All other terms that arise from fifth order vanish because of the reasons discussed above. By numerically[21] performing the integrals in the sum in (B7) we obtain $\xi\approx 0.36$.

When $\langle a\rangle\neq\frac{1}{2}$ the contributions (B4)–(B6) do not vanish, and their evaluation becomes more difficult. As we have argued in Sec. III, however, we might expect that the corrections that result from keeping higher-order terms will lead to only slight (i.e., linear) deviations of $\langle a\rangle$ from $\frac{1}{2}$. We thus must consider the expansions of these terms about this limit. We do this by first introducing the deviation $\lambda=\frac{1}{2}-\langle a\rangle=\langle b\rangle-\frac{1}{2}$, and then taking derivatives with respect to λ. For example, we write

$$z_0(\lambda)=z_0(0)+\lambda b_0$$

where

$$b_0=[\partial z_0(\lambda)/\partial\lambda]_{\lambda=0}$$
$$=2\int_0^\infty t\exp(-t)[I_0^2(t/2)-(2/t)I_0(t/2)I_1(t/2)-I_1^2(t/2)]dt$$
$$=4(1-2/\pi) \tag{B8}$$

so that near $\langle a\rangle=\frac{1}{2}$, $z_0\sim b_0\lambda=\gamma\langle 2a-1\rangle$ with $\gamma=0.73$. From this result, we see that many terms in (B4)–(B6) are of higher order in λ than the first and may therefore be neglected to this order of approximation. Thus, the third-order corrections (B5) may be neglected entirely, along with the first two terms in the fourth-order correction (B6). We now identify the three terms which appear within parentheses in Eq. (3.19) as arising from, respectively, the second-order term (B4), the last two terms of the fourth-order correction (B5), and the nonvanishing term from the fifth-order contribution (B7). The quantities ξ and γ appearing in (3.19) have already been discussed. The quantity β which appears in (3.19) arises from the expansion of the fourth-order corrections around $\langle a \rangle = \frac{1}{2}$ and is given explicitly by

$$\beta = 2 \sum_{\mathbf{n}} (1 + \tfrac{3}{2} b_{\mathbf{n}})[z_{\mathbf{n}}(0)]^2 , \tag{B9}$$

where $z_{\mathbf{n}}(0)$ is given by (B3) with $\langle a \rangle = \langle b \rangle = \frac{1}{2}$, and

$$\begin{aligned} b_{\mathbf{n}} &\equiv [\partial z_{\mathbf{n}}(\lambda)/\partial\lambda]_{\lambda=0} \\ &= 4 \int_0^\infty dt \exp(-2t) t \, [I''_{n_1}(t) I_{n_2}(t) + I_{n_1}(t) I''_{n_2}(t) \\ &\qquad - 2I'_{n_1}(t) I'_{n_2}(t)] \, . \end{aligned} \tag{B10}$$

In (B10) primes denote differentiation with respect to the argument. A numerical evaluation of (B9) leads to the estimate $\beta \approx 1.1$

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