Stochastic transport in disordered systems

Muhammad Sahimi, Barry D. Hughes, L. E. Scriven, and H. Ted Davis

Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, Minnesota 55455
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We develop a theory of stochastic transport in disordered media, starting from a linear master equation with random transition rates. A Green function formalism is employed to reduce the basic equation to a form suitable for the construction of a class of effective medium approximations (EMAs). The lowest order EMA, developed in detail here, corresponds to recent approximations proposed by Odagaki and Lax [Phys. Rev. B **24**, 5284 (1981)], Summerfield [Solid State Commun. **39**, 401 (1981)], and Webman [Phys. Rev. Lett. **47**, 1496 (1981)]. It yields an effective transition rate W_m which can be identified as the memory kernel of a generalized master equation, and used to define an associated continuous-time random walk on a uniform lattice. The long-time behavior of the mean-squared displacement arising from an initially localized state can be found from W_m, as can diffusion constants in any case where the long-time behavior of the system is diffusive. Detailed calculations are presented for seven lattice systems in one, two, and three dimensions, and for a variety of probability density functions $f(w)$ for the transitions rates. For percolation-type densities, i.e., those with only a fraction $p < 1$ of the bonds transmitting, the EMA predicts three distinct kinds of behavior: localization, "fractal" transport with slower than linear growth of the mean-squared displacement, and diffusion in the cases $p < p_c$, $p = p_c$, $p > p_c$, respectively, where p_c is the bond percolation threshold of the lattice. Depending on the form of $f(w)$ near $w = 0$, critical exponents may take values independent of $f(w)$ ("universality") or heavily dependent on $f(w)$ ("nonuniversality").

I. INTRODUCTION

The problem of stochastic transport in disordered systems is important, in view of its relevance to the modeling of a wide variety of phenomena in random media. A partial list of applications includes the migration of localized excitations among guest molecules in a host,[1] hopping transport in amorphous semiconductors,[2] the quantum motion in a solid of an electron, interacting with fixed impurities of a given number density,[3] frequency-dependent conductivity in superionic conductors,[4] electron scavenging in glasses,[5] oxygen transport in nuclear reactor fuels,[6] motion of electrons in liquids,[7] diffusion of nuclear magnetic spin,[8] dispersion in flow through porous media,[9] diffusion through biological tissue,[10] and transport properties of composite materials.[11]

A common point of departure for many theoretical investigations of stochastic transport is the *master equation*

$$\frac{\partial P_i}{\partial t} = \sum_j [W_{ij}P_j(t) - W_{ji}P_i(t)] , \tag{1}$$

which can be viewed as a description of the motion of a particle on a lattice. Here $P_i(t)$ denotes the probability that the particle will be found at site i at time t and W_{ij} is the transition rate between sites i and j, frequently taken to be nonzero only when sites i and j are nearest neighbors. For a master equation description to be appropriate, it is not necessary that the random medium have lattice structure, or that the transport process involve identifiable particles: equations of the form (1) also arise when the problem of diffusion or conduction in a random continuum is discretized by finite difference or finite element means. However, irrespective of the precise physical model, it is necessary to incorporate either or both of the following considerations into the analysis:

(i) the transition rates W_{ij} are random variables;

(ii) the topological structure of the lattice varies randomly with spatial position.

Relatively little work has been done on transport processes in topologically disordered systems.[12] In the main, attention has centered on periodic lattices with random transition rates, i.e., on regular lattices randomly decorated. Even within this restricted context, few exact results have been derived, notable exceptions being confined to one-dimensional systems.[13]

One approach to the problem in two or more dimensions is based upon an effective medium approximation (EMA). Although its application to stochastic transport has been suggested only very recently,[14–16] the basic EMA idea has a long history.[17] It has been extensively used to estimate the effective properties of heterogeneous or composite continua, and a variant has appeared during the last two decades in several areas of solid state and condensed matter physics, where it is often called the coherent potential approximation (CPA).[18] The recent popularity of EMA ideas for lattice systems is largely due to the seminal paper of Kirkpatrick,[19] who used an EMA to describe percolation and (dc) conduction properties of regular lattices of random resistors.

It was realized independently by Summerfield,[14] Webman,[15] and Odagaki and Lax[16] that an EMA could be used to describe stochastic transport governed by the master equation (1), with symmetric transition rates ($W_{ij} = W_{ji}$) which are independent, identically distributed random variables with probability density function $f(w)$. Of particular interest to these authors was the binary distribution

$$f(w) = p\delta(w - w_0) + (1 - p)\delta_*(w) , \tag{2}$$

the prototype for diffusion on a percolating lattice, and the lattices considered were the linear chain, and the three-dimensional simple cubic lattice.[20]

The perspective of Summerfield, Webman, and

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Odagaki and Lax is that of solid state physics, and is reflected in their formalism, which closely parallels coherent potential analyses of condensed matter problems. As an alternative view of the stochastic transport problem, we present a simpler Green function formalism, which leads to clearer derivations of their EMA results and which is more readily generalized to topologically disordered lattices[21] and to the construction of higher order EMA's.[22] We only sketch the possible generalizations here, confining our attention in the main to a systematic investigation of the effects on the lowest order EMA for a periodic lattice of (i) variation in dimensionality and coordination number and (ii) qualitative changes in the structure of the probability density function $f(w)$ for the transition rates. The results given by Summerfield, Webman, and Odagaki and Lax are briefly derived within the present formalism for completeness. We put particular emphasis on the relation between the effective medium approximation, master equations, generalized master equations, and continuous-time random walks, which we use to clarify the nature of qualitatively different types of transport predicted by the EMA.

The paper may be summarized as follows. In Sec. II, we develop a Green function formalism for constructing effective medium approximate solutions of the master equation (1), which applies for any lattice system, whether periodic or topologically disordered. For a periodic lattice, the necessary Green function can be constructed by Fourier analysis (as discussed in the Appendix). We consider in detail the following periodic lattice systems, which exemplify the effect of varying the coordination number z and the dimensionality d of the lattice:

(a) $d=1$: a linear chain ($z=2$),

(b) $d=2$: hexagonal ($z=3$), square ($z=4$), triangular ($z=6$),

(c) $d=3$: diamond ($z=4$), simple cubic ($z=6$), body-centered cubic ($z=8$), face-centered cubic ($z=12$).

For a topologically disordered lattice, such as that generated by a Voronoi tessellation of the plane or of space,[23] the general approach remains valid, provided that the Green function is appropriately interpreted as a random variable, as discussed elsewhere.[21] The Green function formalism of Sec. II emphasizes lattice sites. Its reformulation in Sec. III to emphasize bonds is the point of entry into the systematic construction of EMA's. The simplest EMA (single-bond EMA) is constructed by allowing the transition rate of a selected bond to fluctuate from the characteristic rate (conductivity) W_m of a uniform lattice in which the selected bond is embedded, and deducing a self-consistent value for W_m. This EMA corresponds to the Kirkpatrick[19] EMA for the dc conductivity of a regular lattice of random resistors. Higher order EMA's can be constructed by selecting several bonds, the conductivities of which fluctuate from the uniform value. This idea has been applied by a few authors[24] to the dc conductivity of regular lattices of random resistors, and will be pursued in the present context in a future paper.[25]

At the single bond level, our self-consistency equation for W_m is the same as that derived by Odagaki and Lax, Summerfield and Webman. Before proceeding with the analysis of solutions of this equation for various lattice systems and transition rate distributions, we show in Sec. IV how the EMA value of the effective transition rate W_m can be interpreted as the memory kernel of a generalized master equation, thereby determining the waiting-time distribution $\psi(t)$ for a continuous-time random walk on the lattice. Depending upon the long-time behavior of $\psi(t)$, the mean-squared displacement $\langle R^2(t)\rangle$ arising from an initially localized state can behave as $t\to\infty$ in three qualitatively distinct ways:

(i) classical diffusion or conduction: $\langle R^2(t)\rangle \propto t$, the proportionality constant yielding the diffusion constant and $\langle t\rangle < \infty$, where $\langle t\rangle$ is the mean time between steps in the associated continuous-time random walk;

(ii) nondiffusive conduction: $\langle R^2(t)\rangle \to \infty$, but $\langle R^2(t)\rangle / t \to 0$ and $\langle t\rangle = \infty$;

(iii) localization: $\langle R^2(t)\rangle \to$ constant, and the associated continuous-time random walk may freeze after any step.

The analysis of Sec. V shows that the single-bond EMA predicts all three types of behavior for percolationlike distributions, i.e., distributions for which only a fraction $p<1$ of the bonds are active (have nonzero transfer rate). If the subdistribution of the active bonds satisfies a certain moment condition, we obtain universal behavior, in the sense that certain critical exponents are independent of the distribution. The universality fails if the moment condition is violated. For the binary distribution (2), the behavior of $\langle R^2(t)\rangle$ below and at the percolation threshold serves as a probe of the structure of connected subsets (clusters) of the lattice.[49] Our results are tabulated and interpreted in Sec. VI, where we also suggest extensions of the ideas of the present paper to more complicated systems.

II. SITE GREEN FUNCTION FORMALISM

Introduction of the Laplace Transform

$$\hat{P}_i(\lambda) = \int_0^\infty e^{-\lambda t} P_i(t)\,dt \tag{3}$$

reduces Eq. (1) to an algebraic system

$$\lambda \hat{P}_i(\lambda) - \delta_{i0} = \sum_j [W_{ij}\hat{P}_j(\lambda) - W_{ji}\hat{P}_i(\lambda)] . \tag{4}$$

We restrict our attention to lattices for which $W_{ij} = 0$ unless sites i and j are nearest neighbors. When $W_{ij} = W_{ji}$, as is assumed here,[26] Eq. (4) is equivalent to Kirchhoff's law for an electrical network, with $\hat{P}_j(\lambda)$ corresponding to the voltage at site j and W_{ij} the conductivity of the bond joining sites i and j. The terms $-\delta_{i0}$ and $\lambda\hat{P}_i(\lambda)$ can be interpreted, respectively, as a current source and an electrical connection to ground (with λ a conductivity if it is real, or an admittance if it is complex). In the discussion of the diffusion or conduction problem which follows, we shall frequently resort to electrical circuit terminology, voltage being equivalent to probability or concentration, voltage drop or potential difference to probability or concentration difference, and current to probability or diffusion flux.

To facilitate solution of Eq. (4) we introduce a *reference lattice* with all nearest-neighbor transition rates equal to W^0, and site occupation probabilities $P_i^0(t)$, so that

$$\lambda \hat{P}_i^0(\lambda) - \delta_{i0} = \sum_{j\in\{i\}} W^0[\hat{P}_j^0(\lambda) - \hat{P}_i^0(\lambda)] , \tag{5}$$

where $\{i\}$ denotes the set of nearest neighbors of site i. As is shown in Sec. III, the effective medium approximate solution is constructed by taking $W^0 = W_m(\lambda)$, the effective transition rate of the lattice, and determining $W_m(\lambda)$ in a self-consistent manner. Subtracting Eqs. (4) and (5), we obtain

$$\hat{P}_i(\lambda) - \hat{P}_i^0(\lambda) = \frac{1}{\epsilon} \sum_{j\in\{i\}} \{\Delta_{ij}[\hat{P}_j(\lambda) - \hat{P}_i(\lambda)] + [\hat{P}_j(\lambda) - \hat{P}_i(\lambda)] - [\hat{P}_j^0(\lambda) - \hat{P}_i^0(\lambda)]\} , \tag{6}$$

where $\epsilon \equiv \lambda / W^0$, and $\Delta_{ij} \equiv (W_{ij} - W^0)/W^0$ if i and j are nearest neighbors and zero otherwise. Equation (6) can be rewritten in the form

$$(z_i + \epsilon)[\hat{P}_i(\lambda) - \hat{P}_i^0(\lambda)] - \sum_{j\in\{i\}} [\hat{P}_j(\lambda) - \hat{P}_j^0(\lambda)] = -\sum_{j\in\{i\}} \Delta_{ij}[\hat{P}_i(\lambda) - \hat{P}_j(\lambda)] , \tag{7}$$

where z_i is the coordination number of site i (i.e., the number of nearest neighbors). An associated Green function G is defined by the equation

$$(z_i + \epsilon) G_{ik} - \sum_{j\in\{i\}} G_{jk} = -\delta_{ik} . \tag{8}$$

(G_{ij} is the voltage induced at the lattice site j by unit current injected at site i.) Equation (7) becomes

$$\hat{P}_i(\lambda) = \hat{P}_i^0(\lambda) + \sum_{j,k} G_{ij}\Delta_{jk}[\hat{P}_j(\lambda) - \hat{P}_k(\lambda)] . \tag{9}$$

The analysis so far is exact and applies to any lattice, irrespective of dimensionality or topological structure. For topologically disordered lattices, further progress requires a statistical treatment of the Green function appropriately coupled to the disorder of the lattice.[21] However, for periodic lattices, for which all sites have the same coordination number z, the Green functions can be constructed explicitly by Fourier analysis (see the Appendix).

For a simple cubic lattice in d dimensions ($z = 2d$), which includes as special cases the usual square ($z = 4$) and 3-d simple cubic ($z = 6$) lattices, the construction is particularly straightforward. If we label by d integers $(m_1, m_2, \ldots, m_d)$ the relative position of two sites i and j on the lattice, separated by m_k bond lengths along principal axis k, then provided that $\mathrm{Re}(\epsilon) \geq 0$, if $d \geq 3$, and $\mathrm{Re}(\epsilon) \geq 0$, $\epsilon \neq 0$, if $d = 1$ or 2, a suitable Green function is

$$G_{ij} = G(m_1, \ldots, m_d) = -\frac{1}{2}\int_0^\infty \exp[-\tfrac{1}{2}t(z+\epsilon)] \prod_{i=1}^{d} I_{m_i}(t)\,dt . \tag{10}$$

Here $I_\nu(t)$ denotes the modified Bessel function of order ν. The correctness of Eq. (10) is easily verified by substitution into Eq. (8) and use of the identity

$$I_{\nu-1}(t) + I_{\nu+1}(t) = 2I_\nu'(t).$$

In one or two dimensions, the limit of Eq. (10) as $\epsilon \to 0$ is singular, corresponding to the failure of the Laplace transform $\hat{P}_i(\lambda)$ to converge at $\lambda = 0$, while in three or more dimensions the Green function (10) is continuous at $\epsilon = 0$. As is shown in the Appendix, the singularity or continuity of the Green function as $\epsilon \to 0$ is determined by the dimensionality of the lattice: the Green functions for the hexagonal and triangular lattices diverge at the origin, while those for the face-centered and body-centered cubic lattices and the diamond lattice are continuous. The singular part of the Green function G_{ij} for each of the two-dimensional lattices is independent of the lattice sites i and j,

$$G_{ij} \sim -\frac{A}{4\pi}\ln(1/\epsilon), \quad \text{as } \epsilon \to 0 , \tag{11}$$

where

$$A = \begin{cases} 1, & \text{for the square lattice} \\ 1/\sqrt{3}, & \text{for the triangular lattice} \\ \sqrt{3}, & \text{for the hexagonal lattice} \end{cases} . \tag{12}$$

It will be seen that the construction of the single-bond EMA in Sec. III involves only linear combinations of the G_{ij} which define functions continuous at the origin.

The continuity of the Green function in three or more dimensions means that the steady state injection problem

$$zG_{ik} - \sum_{j\in\{i\}} G_{jk} = -\delta_{ik} \tag{13}$$

can be treated as the small λ limit of the general time-dependent problem defined by Eq. (9). However, for one or two dimensions, one should in principle treat the steady state problem separately. A Green function for Eq. (13) can be constructed from the Green function of Eq. (8) by subtracting G_{kk} and letting $\epsilon \to 0$. For example, the time-dependent Green function for the square lattice

$$G(m_1, m_2) = -\frac{1}{2}\int_0^\infty \exp(-2t - \tfrac{1}{2}\epsilon t)\, I_{m_1}(t)\, I_{m_2}(t)\,dt \tag{14}$$

can be replaced, for $\epsilon = 0$, by the Green function

$$G(m_1, m_2) = \frac{1}{2}\int_0^\infty \exp(-2t)[I_0^2(t) - I_{m_1}(t)I_{m_2}(t)]\,dt . \tag{15}$$

The qualitative difference between the cases $d \leq 2$ and $d \geq 3$ in the present context reflects a characteristic difference between stochastic processes in low and higher dimensional systems which is well known in other contexts. Pólya's random walk[27] on a d-dimensional cubic lattice provides an example. The Green function for Pólya's walk is, apart from minor notational differences, given by Eq. (10). For sufficiently low dimensionality ($d = 1$ or $d = 2$), the divergence of the Green function at $\epsilon = 0$ establishes that the walker must return to the origin, while its finiteness for $d \geq 3$ ensures that the walk is transient, i.e., the walker need not return to the origin and will ultimately escape to infinity.

III. BOND GREEN FUNCTION FORMALISM AND THE SINGLE-BOND EMA

The electrical circuit analogy suggests that it may be useful to work in terms of the difference in $\hat{P}$ between neighboring sites, rather than the values of $\hat{P}$ at the sites themselves. Let $\hat{Q}_{ij}(\lambda)$ denote the "voltage drop" across the nearest-neighbor link i–j, i.e.,

$$\hat{Q}_{ij}(\lambda) \equiv \hat{P}_i(\lambda) - \hat{P}_j(\lambda) , \tag{16}$$

so that $\hat{Q}_{ij}(\lambda) = -\hat{Q}_{ji}(\lambda)$. Then Eq. (9) becomes

$$\hat{Q}_{ij}(\lambda) = \hat{Q}^0_{ij}(\lambda) + \sum_l \sum_k \hat{Q}_{lk}(\lambda)\Delta_{lk}(G_{il} - G_{jl}) , \tag{17}$$

so that

$$\hat{Q}_{ij}(\lambda) = \hat{Q}^0_{ij}(\lambda) + \sum_{[lk]} \Delta_{lk}\hat{Q}_{lk}(\lambda)(G_{il} + G_{jk} - G_{jl} - G_{ik}) , \tag{18}$$

where $[lk]$ means that the bond connecting nearest-neighbor sites l and k is counted only once. We denote bonds with Greek letters and assign directions to them and let

$$\gamma_{\alpha\beta} \equiv (G_{il} + G_{jk}) - (G_{jl} + G_{ik}) \tag{19}$$

be a bond–bond Green function,[28] where i and l (j and k) are lattice sites with tails (heads) of arrows on bond α and β respectively. Unlike the site–site Green function G_{ik}, the bond–bond Green function $\gamma_{\alpha\beta}$ is not singular in one or two dimensions when $\epsilon \to 0$. Equation (18) can be written as

$$\hat{Q}_\alpha(\lambda) = \hat{Q}^0_\alpha(\lambda) + \sum_\beta \Delta_\beta \gamma_{\alpha\beta}\hat{Q}_\beta(\lambda) . \tag{20}$$

The effective transition rate (conductivity) for the lattice, denoted by W_m, can be defined by the following algorithm. Define the reference lattice such that $W_m = W^0$ and solve Eq. (20) for an arbitrary set of individual transition rates, i.e., for arbitrary Δ_β. Select any bond α and require that

$$\langle \hat{Q}_\alpha - \hat{Q}^0_\alpha \rangle_\infty = 0 , \tag{21}$$

where $\langle\ \rangle_\infty$ denotes the average over all possible transition rates of all bonds. The condition (21) requires that the fluctuation in the voltage drop across bond α from its value for the reference lattice should vanish on the average and leads to a self-consistent determination of W_m. (The W_m so obtained is a function of λ, and though we refer to it here loosely as an effective transition *rate*, it has the significance of a *memory kernel* in a *generalized master equation*, as discussed below.)

Such an algorithm cannot be implemented in practice. As an approximation, however, one may assign to all but a finite number of bonds in the lattice the transition rate W_m (so that $\Delta_\beta \neq 0$ only for a finite number of bonds), and proceed as above, now averaging over the conductivities of a finite number of bonds to determine W_m self-consistently. To obtain rapid convergence with increasing cluster size to the infinite system result, it is important to choose a suitably symmetrical cluster of bonds whose transition rates are to be allowed to fluctuate from W_m. These matters are discussed in detail elsewhere.[22] For the present, we consider only the simplest case, in which only a single bond (α) has transition rate W differing from W_m. Equation (20) reduces to

$$\hat{Q}_\alpha(\lambda) = \frac{1}{1 - \gamma_{\alpha\alpha}\Delta_\alpha}\hat{Q}^0_\alpha(\lambda) . \tag{22}$$

We now average over all possible values of W, requiring that $\langle \hat{Q}_\alpha(\lambda) \rangle = \hat{Q}^0_\alpha(\lambda)$, i.e., the voltage fluctuation induced by W differing from W_m is to vanish on the average. A self-consistent determination of W_m results:

$$\left\langle \frac{1}{1 - \gamma_{\alpha\alpha}\Delta_\alpha} \right\rangle = 1 . \tag{23}$$

(The same equation is obtained here if a different self-consistency condition is imposed, viz., that the fluctuation in current is to vanish: $\langle W_\alpha Q_\alpha \rangle = W_m \hat{Q}^0_\alpha$.) It remains only to specify the lattice (to determine $\gamma_{\alpha\alpha}$) and the transition rate distribution $f(w)$. For the periodic lattices considered in the present paper, it may be easily shown [using Eqs. (19) and (A23)] that

$$\gamma_{\alpha\alpha} = -2/z + (2\epsilon/z)\,\mathcal{G}(\epsilon) , \tag{24}$$

where

$$\mathcal{G}(\epsilon) = -G_{ii} > 0 , \tag{25}$$

with i the site at which the right-hand side of Eq. (8) is nonzero, i.e., the site of current injection, in the electrical terminology. Equation (23) has been derived recently by Webman,[15] Summerfield,[14] and Odagaki and Lax[16] using different techniques.[52]

IV. RELATION BETWEEN THE EMA, GENERALIZED MASTER EQUATIONS, AND CONTINUOUS-TIME RANDOM WALKS

Before examining particular lattices and distributions, we examine which properties of $W_m(\lambda)$ are needed to characterize the transport process. Replacing W_{ij} in the Laplace transformed master equation (4) by $W_m(\lambda)$ gives

$$\lambda\hat{P}_i(\lambda) - \delta_{i0} = W_m(\lambda) \sum_{j\in\{i\}} [\hat{P}_j(\lambda) - \hat{P}_i(\lambda)] \tag{26}$$

and, inverting the Laplace transform, we find

$$\frac{\partial P_i}{\partial t} = \sum_{j\in\{i\}} \int_0^t \check{W}_m(t-\tau)\,[P_j(\tau) - P_i(\tau)]\,d\tau . \tag{27}$$

Thus, if we can determine the effective transition rate $W_m(\lambda)$ exactly, we can replace the *master equation* (1) for a lattice with random transition rates by the *generalized master equation* (27) for a lattice with uniform memory kernels $\check{W}_m(t) = \mathcal{L}^{-1} W_m(\lambda)$. In practice, since $W_m(\lambda)$ is determined here only implicitly through Eq. (21), an equation which we can usually only solve approximately (via the EMA), we are only able to derive an approximate generalized master equation.[31] Before we address the specific predictions of the EMA-based generalized master equation, we recall that the kinds of stochastic behavior which can be described by equations of the form (27) are more varied than those describable by the memoryless equation

$$\frac{\partial P_i}{\partial t} = \text{const.} \sum_{j\in\{i\}} [P_j(t) - P_i(t)] . \tag{28}$$

It has been shown by Kenkre *et al.*[29] that a general-

ized master equation for a periodic lattice, of the form

$$\frac{\partial}{\partial t}P(\mathbf{l},t) = \int_0^t d\tau\,\phi(t-\tau)\sum_{\mathbf{l}'}\{p(\mathbf{l}-\mathbf{l}')P(\mathbf{l}',\tau)-p(\mathbf{l}'-\mathbf{l})P(\mathbf{l},\tau)\}, \tag{29}$$

can be put into a one-to-one correspondence with a continuous-time random walk[30] on the same lattice, with transition probability $p(\mathbf{l}-\mathbf{l}')$ for a displacement from site $\mathbf{l}'$ to site $\mathbf{l}$, and a waiting time density $\psi(t)$ describing the time between steps.[31] The relation between $\psi(t)$ and the time-dependent part $\phi(t)$ of the memory kernel in the generalized master equation is most simply expressed in Laplace transform space:

$$\hat{\psi}(\lambda)=\frac{\hat{\phi}(\lambda)}{\lambda+\hat{\phi}(\lambda)}\ . \tag{30}$$

If we restrict our attention to periodic lattices, with lattice sites defined by position vectors $\mathbf{l}$ and motion commencing from the site $\mathbf{l}=\mathbf{0}$, we have

$$\lambda\hat{P}(\mathbf{l},\lambda)-\delta_{\mathbf{l},\mathbf{0}}=W_m(\lambda)\sum_{\mathbf{l}'\in\{\mathbf{l}\}}\{\hat{P}(\mathbf{l}',\lambda)-\hat{P}(\mathbf{l},\lambda)\}\ . \tag{31}$$

We may choose to analyze this equation as a generalized master equation, or (in view of the result of Kenkre *et al.*[29]) we may consider the equivalent continuous-time random walk, characterized by a waiting time density $\psi(t)$, with

$$\hat{\psi}(\lambda)=\frac{z\,W_m(\lambda)}{\lambda+z\,W_m(\lambda)} \tag{32}$$

(where z is the coordination number of the lattice). We now apply a discrete Fourier transform to Eq. (31), with

$$\tilde{P}(\mathbf{k},\lambda)=\sum_{\mathbf{l}}\exp(i\mathbf{k}\cdot\mathbf{l})\hat{P}(\mathbf{l},\lambda)\ , \tag{33}$$

so that

$$\tilde{P}(\mathbf{k},\lambda)=\{\lambda+z\,W_m(\lambda)[1-\Lambda(\mathbf{k})]\}^{-1}\ , \tag{34}$$

where

$$\Lambda(\mathbf{k})=(1/z)\sum_{\mathbf{l}'\in\{\mathbf{l}\}}\exp[i\mathbf{k}\cdot(\mathbf{l}'-\mathbf{l})] \tag{35}$$

is the structure function of the lattice[27] and z is the coordination number. The Laplace transform of the mean-squared displacement at time t is

$$\mathcal{L}\{\langle R^2(t)\rangle;\ t\to\lambda\}=\sum_{\mathbf{l}}\mathbf{l}^2\hat{P}(\mathbf{l},\lambda) \tag{36}$$

$$=-\nabla_{\mathbf{k}}^2\tilde{P}(\mathbf{k},\lambda)\big|_{\mathbf{k}=0}\ , \tag{37}$$

and if the bonds of the lattice all have unit length we easily see that $\nabla_{\mathbf{k}}\Lambda(\mathbf{k})=0$ and $-\nabla_{\mathbf{k}}^2\Lambda(\mathbf{k})=1$ at $\mathbf{k}=\mathbf{0}$. It follows that

$$\langle R^2(t)\rangle=\mathcal{L}^{-1}\{(z/\lambda^2)W_m\}\ . \tag{38}$$

The long-time behavior of $\langle R^2(t)\rangle$ can be inferred from the behavior of its Laplace transform in the neighborhood of the origin[32] and will therefore depend on the behavior of $W_m(\lambda)$ as $\lambda\to 0^+$. In view of Eq. (32), the small λ behavior of $W_m(\lambda)$ determines the long-time behavior of the waiting time density for the associated continuous-time random walk. We consider three distinct cases for the behavior of $W_m(\lambda)$ as $\lambda\to 0^+$:

(i) $W_m(\lambda)\to C$, a constant. Since $\mathcal{L}^{-1}(\lambda^{-2})=t$, we then have $\langle R^2(t)\rangle\sim zCt$ as $t\to\infty$, and so there is macroscopic transport or conduction. Moreover, this transport is diffusive in character for sufficiently large times, since the fundamental solution of the continuum diffusion equation $\partial p/\partial t=D\nabla^2 p$ in d dimensions gives $\langle R^2(t)\rangle=2dDt$, and we may identify the diffusion constant as

$$D=(z/2d)C\ . \tag{39}$$

Any memory kernel for which $\int_0^\infty \check{W}_m(t)\,dt=C$ will give $W_m(\lambda)\to C$ as $\lambda\to 0^+$ [including the cases $\check{W}_m(t)=C\alpha\exp(-\alpha t)$ and $\check{W}_m(t)=C\delta_+(t)$; in the latter case the generalized master equation (27) reduces to the usual master equation (28)].

(ii) $W_m(\lambda)\sim C\lambda$, C constant. Since $\mathcal{L}^{-1}(\lambda^{-1})=1$, we have $\langle R^2(t)\rangle\to zC$. In this case there is no macroscopic conduction and the motion is confined for all times to a finite region of the lattice. It is not possible to find a non-negative continuous function $\check{W}_m(t)$ for which $W_m(\lambda)\sim C\lambda$ as $\lambda\to 0^+$. However, we are able to generate this asymptotic behavior from oscillating memory kernels such as

$$\check{W}_m(t)=C\omega^2\cos\omega t\ , \tag{40}$$

from memory kernels which change sign a finite number of times and decay at large times, such as

$$\check{W}_m(t)=C\alpha^2(1-\alpha t)\exp(-\alpha t)\ , \tag{41}$$

and from any memory kernel of the form $\check{W}_m(t)=f'(t)+f(t)$, where $f(t)\to 0$ as $t\to\infty$ and

$$f(0^+)=\int_0^\infty f(t)dt=C\ . \tag{42}$$

(iii) $W_m(\lambda)\to 0$, but $W_m(\lambda)/\lambda\to\infty$. In this case, we have $\langle R^2(t)\rangle\to\infty$, so that there is conduction, but this conduction is not diffusive in character, since $\langle R^2(t)\rangle$ grows with time more slowly than t. If we attempt to write $\langle R^2(t)\rangle\sim 2dDt$, we are forced to conclude that the "diffusion constant" D is a function of time which decays at long times. We consider an example:

$$W_m(\lambda)\sim C\lambda^\alpha,\quad 0<\alpha<1\ . \tag{43}$$

Since

$$\mathcal{L}^{-1}\{\lambda^{\alpha-2}\}=\Gamma(2-\alpha)^{-1}t^{1-\alpha} \tag{44}$$

(where Γ denotes the usual gamma function), we find that

$$\langle R^2(t)\rangle\sim\{Cz/\Gamma(2-\alpha)\}t^{1-\alpha}\ . \tag{45}$$

A memory kernel yielding the value $\alpha=\frac{1}{2}$ may be inferred from the Laplace transform pair[33]

$$W_m(t)\propto\frac{1}{\sqrt{\pi t}}-\frac{2a}{\sqrt{\pi}}\exp(-a^2t)\int_0^{a\sqrt{t}}\exp(\tau^2)\,d\tau=\mathcal{L}^{-1}\left\{\frac{\sqrt{\lambda}}{a^2+\lambda}\right\}. \tag{46}$$

A clear physical interpretation of these three distinct cases is possible, using the correspondence between generalized master equations and continuous-time random walks. It follows from Eq. (32) that:

$$\int_0^\infty dt\,\psi(t)=\lim_{\lambda\to 0^+}\hat{\psi}(\lambda)=\begin{cases}1\ , & \text{in cases (i) and (iii)}\\ \dfrac{zC}{1+zC}\ , & \text{in case (ii)}\end{cases}. \tag{47}$$

In cases (i) and (iii), the random walk will never stop, since if the walker arrives at lattice site 1, he is certain to leave it at some subsequent time. On the other hand, in case (ii), there is a finite probability $(1+zC)^{-1}$ that a walker arriving at a site 1 never leaves it. Indeed, the probability that precisely n steps are taken in the entire history of the random walk is $(zC)^n(1+C)^{-n-1}$, a rapidly decaying function of n, and the mean number of steps taken is

$$\sum_{n=1}^{\infty} n(zC)^n(1+zC)^{-n-1} = zC \ . \tag{48}$$

There is a striking difference between the character of $\psi(t)$ in cases (i) and (iii). In case (i),

$$\hat{\psi}(\lambda) \sim 1-(zC)^{-1}\lambda + \text{higher terms}, \tag{49}$$

while in case (iii), if $W_m(\lambda) \sim C\lambda^{\alpha}$ then

$$\hat{\psi}(\lambda) \sim 1-(zC)^{-1}\lambda^{1-\alpha} + \text{higher terms} \ . \tag{50}$$

It can be shown for arbitrary $\psi(t)$ that if the mean time between jumps

$$\langle t \rangle = \int_0^{\infty} t\psi(t)\,dt \tag{51}$$

is finite, then

$$\hat{\psi}(\lambda) = 1-\langle t \rangle \lambda + \text{higher terms}, \tag{52}$$

so that in case (i), $\langle t \rangle$ is finite, while in case (iii) $\langle t \rangle = \infty$. It is readily established[34] that a waiting-time distribution $\psi(t)$ for which

$$\psi(t) \sim \{zC|\Gamma(\alpha-1)|\}^{-1} t^{\alpha-2} \ , \quad \text{as } t \to \infty \ (0<\alpha<1) \tag{53}$$

gives rise to the asymptotic behavior specified by Eq. (50). Such "long-tailed" waiting time distributions have been used in continuous-time random walk models of charge transport in xerographic films[2] and can be related[35] to sets of fractal dimension[36] and real-space renormalization group transformations.[37] We shall call transport described by such distributions "fractal". It should be noted that although the occurrence of an infinite mean time between jumps in case (iii) may be interpreted as a kind of freezing of motion, it does not represent genuine localization. The median time between jumps is finite. We now show that within the single-bond EMA all three types of stochastic motion (diffusive, terminating, and fractal) arise, depending on the dimensionality of the system and the structure of the transition rate distribution $f(w)$.

V. PREDICTIONS OF THE SINGLE-BOND EMA FOR PERIODIC LATTICES

For any distribution $f(w)$ of transition rates, the single-bond EMA is, from Eqs. (23) and (24),

$$\int_0^{\infty} \frac{f(w)\,dw}{1-p_c+p_c\epsilon\,\mathcal{G}(\epsilon)+p_c\{1-\epsilon\,\mathcal{G}(\epsilon)\}(w/W_m)} = 1 \ , \tag{54}$$

where $\epsilon \equiv \lambda/W_m(\lambda)$ and we have defined

$$p_c \equiv 2/z \ . \tag{55}$$

The quantity p_c is the *single-bond effective medium approximation* to the bond percolation threshold[19] of a lattice with coordination number z. As we shall shortly demonstrate, if the inactive bond fraction

$$1-p = \lim_{\eta \to 0} \int_0^{\eta} f(w)\,dw \tag{56}$$

is nonzero, i.e.,

$$f(w) = (1-p)\,\delta_+(w) + ph(w) \ , \tag{57}$$

where $h(w)$ has no generalized function component at the origin, then qualitatively different behavior is obtained for the three regimes $0<p<p_c$, $p=p_c$, $p_c<p<1$. The quantity p represents the relative abundance of bonds which are active. We call Eq. (57) a *percolation-like* distribution because it leads [as does the binary distribution (2)] to a percolation threshold in dc conduction. Using the EMA, Kogut and Straley[38] have considered the dc conductivity of regular lattices of random resistors, with a conductivity distribution of the form (57), and found that there is a striking difference in behavior of the effective conductivity when p is near p_c depending on whether

$$h_{-1} \equiv \int_0^{\infty} \frac{h(w)\,dw}{w} \tag{58}$$

is finite or infinite. If h_{-1} is finite, "universal" results are obtained, i.e., certain critical exponents take values independent of $h(w)$. However, if $h_{-1}=\infty$, the critical exponents depend heavily on the behavior of $h(w)$ as $w \to 0^+$. This result has also been obtained for dc conductivity, using renormalization group techniques, by Ben-Mizrahi and Bergman.[39] We shall exhibit analogous results for stochastic transport.

For a distribution with all bonds active (i.e., $p=1$), we see that so long as $z>2$ (i.e., in two or more dimensions),[53] it is a direct consequence of Eq. (54) that $W_m(\lambda) \to W_m(0)$, a constant, as $\lambda \to 0^+$, where $W_m(0)$ is the solution of

$$\int_0^{\infty} \frac{f(w)\,dw}{1-p_c+p_c w/W_m(0)} = 1 \ . \tag{59}$$

Thus, if the lattice has dimension $d \geq 2$, for any transition rate distribution with all bonds active, the long-time behavior of the system is diffusive, with the diffusion constant D determined by Eq. (59) and the relation $D = (z/2d)W_m(0)$ [Eq. (39)].

We now turn to a more detailed analysis of the cases in which diffusive behavior is not guaranteed on general grounds: the one-dimensional lattice, and percolation-like transition rate probability densities in higher dimensions. In the analysis we frequently employ Tauberian theorems[32] for the determination of the long-time behavior of functions from the small λ behavior of their Laplace transforms. We also employ well-known methods for the asymptotic solution of transcendental equations.[40] Most of the details are straightforward, and are consequently omitted.

A. The linear chain (d = 1)

For a linear chain, $p_c = 1$ and Eq. (54) reduces to

$$\int_0^{\infty} \frac{f(w)\,dw}{\epsilon\,\mathcal{G}(\epsilon)+[1-\epsilon\,\mathcal{G}(\epsilon)](w/W_m)} = 1 \ , \tag{60}$$

where [see the Appendix]

$$\mathcal{G}(\epsilon)=\epsilon^{-1/2}[4+\epsilon]^{-1/2}\ . \tag{61}$$

We consider first the case when all bonds are active ($p=1$). So long as

$$f_{-1}=\int_0^\infty \frac{f(w)\,dw}{w} \tag{62}$$

is finite, then as $\lambda\to 0^+$

$$W_m(\lambda)\to W_m(0)=(1/f_{-1})\ , \tag{63}$$

and we have diffusion as $t\to\infty$, with diffusion constant $D=(1/f_{-1})$. On the other hand, if $f_{-1}=\infty$, we have as $\lambda\to 0^+$

$$W_m(\lambda)\int_0^\infty \frac{f(w)\,dw}{\frac{1}{2}\lambda^{1/2}W_m(\lambda)^{1/2}+w}\sim 1\ . \tag{64}$$

If $f(w)\to f(0)\,[0<f(0)<\infty]$ as $w\to 0^+$, we deduce from Eq. (64) that

$$f(0)\,W_m(\lambda)\ln\{2\lambda^{-1/2}W_m(\lambda)^{-1/2}\}\sim 1 \tag{65}$$

and so

$$W_m(\lambda)\sim 2\{f(0)\ln(\lambda^{-1})\}^{-1}\ . \tag{66}$$

In this case the system evolves nondiffusively:

$$\langle R^2(t)\rangle\sim\{2/f(0)\}t/\ln t\ , \tag{67}$$

and the mean time between steps for the associated continuous-time random walk is infinite [the integral (51) being logarithmically divergent]. To deduce Eq. (67) from Eq. (66) we have used Eq. (38) and a Tauberian theorem. If $f(w)\sim cw^{-\alpha}(0<\alpha<1)$ as $w\to 0^+$, we find that

$$W_m(\lambda)\sim\left\{\frac{\sin\pi\alpha}{2^\alpha c\pi}\right\}^{2/(2-\alpha)}\lambda^{\alpha/(2-\alpha)}\ , \tag{68}$$

and

$$\langle R^2(t)\rangle\propto t^{(2-2\alpha)/(2-\alpha)}\ . \tag{69}$$

Equations (67) and (69) have been given previously by Alexander *et al.*[41] Their analysis establishes that these asymptotic forms are exact. Odagaki and Lax[42] have previously presented some of the above results in their CPA analysis of ac conductivity of random linear chains. We conclude that the system is diffusive [$\langle R^2(t)\rangle/t\to$ constant] only if $f(w)$ vanishes sufficiently rapidly as $w\to 0^+$, with fractal transport occurring otherwise.

If $p<1$, i.e., if a fraction of the bonds are inactive, we find in place of Eq. (60), the equation

$$p\int_0^\infty \frac{h(w)\,dw}{\epsilon\,\mathcal{G}(\epsilon)+[1-\epsilon\,\mathcal{G}(\epsilon)](w/W_m)}=\frac{p-1+\epsilon\,\mathcal{G}(\epsilon)}{\epsilon\,\mathcal{G}(\epsilon)}\ , \tag{70}$$

with $h(w)$ defined by Eq. (57). There can be no long-range transport, since the existence of a single inactive bond "breaks the circuit". Motion must therefore be localized and we recall that this requires $W_m(\lambda)\sim\kappa^{-1}\lambda$, as $\lambda\to 0^+$ (with κ constant). We insert this asymptotic form into Eq. (70) and obtain

$$p\kappa^{-1}\lambda\int_0^\infty \frac{h(w)dw}{\lambda\mathcal{G}(\kappa)+[1-\kappa\mathcal{G}(\kappa)]w}\sim\frac{p-1+\kappa\,\mathcal{G}(\kappa)}{\kappa\,\mathcal{G}(\kappa)}\ . \tag{71}$$

For any distribution $h(w)$, the left-hand side of Eq. (71) approaches zero as $\lambda\to 0^+$, and so the right-hand side must vanish identically. This gives an equation for κ:

$$\kappa\,\mathcal{G}(\kappa)=1-p\ . \tag{72}$$

This equation may be solved exactly, giving

$$W_m(\lambda)\sim\tfrac{1}{4}p(2-p)(1-p)^{-2}\lambda \tag{73}$$

and

$$\lim_{t\to\infty}\langle R^2(t)\rangle=\tfrac{1}{2}p(2-p)(1-p)^{-2}\ . \tag{74}$$

These results have been derived previously for the binary distribution (2) by Odagaki and Lax.[16] We have shown that they continue to hold for a general distribution $h(w)$ of the transition rates of active bonds. This is to be expected, since $\lim_{t\to\infty}\langle R^2(t)\rangle=\frac{1}{2}p(2-p)(1-p)^{-2}$ reflects the mean size of clusters (subsets of the lattice connected by active bonds), which is a purely topological property. However, if $h_{-1}=\infty$, the first corrections to Eqs. (73) and (74) will be heavily distribution dependent.

B. Percolationlike distributions in two and three dimensions

It has been shown above that in two or more dimensions, the ultimate behavior of the system is diffusive if the distribution of transition rate is nonpercolative (i.e., $p=1$). We now examine a general percolationlike distribution of the form (57), with $p<1$. Equation (54) becomes

$$p\int_0^\infty \frac{h(w)\,dw}{1-p_c+p_c\epsilon\,\mathcal{G}(\epsilon)+p_c\{1-\epsilon\,\mathcal{G}(\epsilon)\}(w/W_m)}$$
$$=\frac{p-p_c+\epsilon p_c\,\mathcal{G}(\epsilon)}{1-p_c+\epsilon p_c\,\mathcal{G}(\epsilon)}\ . \tag{75}$$

We pose the general question: under what circumstances will the three distinct types of transport discussed in Sec. IV be predicted by Eq. (75)? Consider first the case of *diffusive transport*, where $W_m(\lambda)\to W_m(0)>0$ as $\lambda\to 0^+$. Since

$$\mathcal{G}(\epsilon)\sim\begin{cases}(A/4\pi)[\ln(1/\epsilon)], & \text{in two dimensions,}\\ \text{const.}, & \text{in three dimensions,}\end{cases}$$
$$\text{as } \epsilon\to 0^+\ , \tag{76}$$

we see that as $\lambda\to 0^+$ [implying that $\epsilon\sim\lambda/W_m(0)$] Eq. (75) yields

$$W_m(0)(p/p_c)\int_0^\infty \frac{h(w)\,dw}{(1-p_c)W_m(0)/p_c+w}=\frac{p-p_c}{1-p_c}\ . \tag{77}$$

Solutions of Eq. (77) with $W_m(0)>0$ are possible if and only if $p>p_c$. We see, therefore, that diffusion is obtained after sufficiently long times only above the percolation threshold. The value of $W_m(0)$ (and so of the diffusion constant) decreases as $p\to p_c$ from above, a property which we shall explore in more detail below for the binary distribution. If h_{-1}, defined by Eq. (58), is finite, then

$$W_m(0)\sim\frac{p-p_c}{h_{-1}(1-p_c)},\quad \text{as } p\to p_c^+\ , \tag{78}$$

whereas if $h_{-1}=\infty$, nonuniversal behavior occurs as $p\to p_c^+$, the asymptotic form of $W_m(0)$ depending on the behavior of $h(w)$ near $w=0$. Arguments similar to

those used above in finding the λ dependence of $W_m(\lambda)$ in one dimension may be used to show that:

(i) $h(w) \to h_0 > 0$ implies

$$W_m(0) \sim \{h_0(1-p_c)\}^{-1}(p-p_c)/\ln\{[p-p_c]^{-1}\}\,; \tag{79}$$

(ii) $h(w) \sim cw^{-\alpha}$ $(0<\alpha<1)$ implies

$$W_m(0) \sim \left\{\frac{\sin(\pi\alpha)}{c\pi p_c^{\alpha}}\right\}^{1/(1-\alpha)} \frac{(p-p_c)^{1/(1-\alpha)}}{(1-p_c)}\,. \tag{80}$$

We consider next the case of *localization*, i.e., $\langle R^2(t)\rangle$ bounded as $t\to\infty$, or in the continuous-time random walk interpretation $\int_0^\infty \psi(t)\,dt<1$. This requires $W_m(\lambda)\propto\lambda$, as $\lambda\to 0^+$. To seek such solutions, we write

$$W_m(\lambda) \sim \kappa^{-1}\lambda, \text{ as } \lambda \to 0^+\,. \tag{81}$$

To determine κ, we note that to leading order Eq. (75) becomes

$$\lambda(p/p_c)\int_0^\infty \frac{h(w)\,dw}{(\lambda/p_c)[1-p_c+p_c\kappa\,\mathcal{S}(\kappa)]+\kappa[1-\kappa\,\mathcal{S}(\kappa)]w} \sim \frac{p-p_c+p_c\kappa\,\mathcal{S}(\kappa)}{1-p_c+p_c\kappa\,\mathcal{S}(\kappa)}\,, \tag{82}$$

and for the asymptotic behavior (81) to be obtained the right-hand side of Eq. (82) must vanish identically, i.e.,

$$\kappa\,\mathcal{S}(\kappa) = (1-p/p_c)\,. \tag{83}$$

This equation has positive solutions only below the percolation threshold. We cannot solve Eq. (83) for κ explicitly, but the asymptotic behavior of κ as $p\to p_c^-$ is easily obtained:

(a) in two dimensions, $\mathcal{S}(\kappa)\sim(A/4\pi)\ln(1/\kappa)$, as $\kappa\to 0$, so that

$$W_m(\lambda) \sim (A/4\pi)(1-p/p_c)^{-1}\ln\{(1-p/p_c)^{-1}\}\lambda\,; \tag{84}$$

(b) in three dimensions, $\mathcal{S}(\kappa)\to\mathcal{S}(0)<\infty$, as $\kappa\to 0$, so that

$$W_m(\lambda) \sim \mathcal{S}(0)\,[1-p/p_c]^{-1}\lambda\,. \tag{85}$$

As in one dimension, we find leading order results independent of the distribution $h(w)$ of active bond transition rates. In particular, close to the percolation threshold,

$$\lim_{t\to\infty}\langle R^2(t)\rangle \sim (zA/4\pi)(1-p/p_c)^{-1}\{\ln(1-p/p_c)^{-1}\} \tag{86}$$

in two dimensions, and

$$\lim_{t\to\infty}\langle R^2(t)\rangle \sim z\,\mathcal{S}(0)(1-p/p_c)^{-1} \tag{87}$$

in three dimensions.

We have shown that for percolation-type distributions in two or three dimensions, the single-bond EMA predicts localization if and only if $p<p_c$, and diffusion if and only if $p>p_c$. As we now show, the remaining type of stochastic behavior, *fractal transport*, is predicted at the percolation threshold ($p=p_c$). It may be recalled from Sec. IV that for fractal transport $W_m(\lambda)\to 0$, as $\lambda\to 0^+$ with $\epsilon=\lambda/W_m(\lambda)\to 0$, and $\langle R^2(t)\rangle$ grows with time more slowly than linearly, while the waiting time distribution $\psi(t)$ for the associated continuous-time random walk is long tailed, ($\langle t\rangle=\infty$). Setting $p=p_c$ in Eq. (75), we find as $\lambda\to 0^+$,

$$W_m(\lambda)\int_0^\infty \frac{h(w)\,dw}{(1-p_c)W_m(\lambda)/p_c+w} \sim \frac{\{p_c\lambda/W_m(\lambda)\}\,\mathcal{S}[\lambda/W_m(\lambda)]}{1-p_c}\,. \tag{88}$$

If h_{-1} is finite, we obtain from Eq. (88),

$$W_m(\lambda)^2 \sim \frac{p_c\lambda\,\mathcal{S}[\lambda/W_m(\lambda)]}{(1-p_c)h_{-1}}\,, \tag{89}$$

and we have universal behavior, i.e., qualitative behavior determined by the dimensionality of the system, independent of the structure of $h(w)$. In three (or higher) dimensions, $\mathcal{S}(0)$ is finite, and we obtain

$$W_m(\lambda) \sim \left\{\frac{p_c\,\mathcal{S}(0)}{(1-p_c)h_{-1}}\right\}^{1/2}\lambda^{1/2}\,, \tag{90}$$

and so at large times

$$\langle R^2(t)\rangle \sim \frac{2z}{\pi^{1/2}}\left\{\frac{p_c\,\mathcal{S}(0)}{(1-p_c)h_{-1}}\right\}^{1/2} t^{1/2}\,, \tag{91}$$

and the associated continuous-time random walk has a waiting time distribution

$$\psi(t) \propto t^{-3/2}\,, \quad t\to\infty\,. \tag{92}$$

In two dimensions, using Eq. (11) we see that

$$\mathcal{S}(\lambda/W_m(\lambda)) \sim (A/4\pi)\ln(W_m(\lambda)/\lambda) \tag{93}$$

and Eq. (89) may be solved asymptotically using standard techniques[40] to give

$$W_m(\lambda) \sim \left\{\frac{Ap_c\ln\lambda}{8\pi(1-p_c)h_{-1}}\right\}^{1/2}\lambda^{1/2}\,, \tag{94}$$

$$\langle R^2(t)\rangle \sim \frac{z}{\pi}\left\{\frac{Ap_c\ln t}{2(1-p_c)h_{-1}}\right\}^{1/2} t^{1/2}\,, \tag{95}$$

and

$$\psi(t) \propto t^{-3/2}(\ln t)^{1/2}\,. \tag{96}$$

If $h(w)\to h_0>0$, as $w\to 0^+$, Eq. (88) gives

$$h_0 W_m(\lambda)\ln\{W_m(\lambda)^{-1}\} \sim \frac{\{p_c\lambda/W_m(\lambda)\}\,\mathcal{S}(\lambda/W_m(\lambda))\}}{1-p_c} \tag{97}$$

and so

$$W_m(\lambda)^2\ln\{W_m(\lambda)^{-1}\} \sim \begin{cases} \dfrac{Ap_c\lambda\ln(W_m/\lambda)}{4\pi h_0(1-p_c)}\,, & \text{in two dimensions} \quad (98)\\[2ex] \dfrac{\mathcal{S}(0)p_c\lambda}{h_0(1-p_c)}\,, & \text{in three dimensions} \quad (99)\end{cases}$$

and we find weakly nonuniversal behavior

$$W_m(\lambda) \sim \left\{\frac{Ap_c\lambda}{4\pi h_0(1-p_c)}\right\}^{1/2} \tag{100}$$

in two dimensions, and

$$W_m(\lambda) \sim \left\{\frac{2\,\mathcal{S}(0)p_c\lambda}{h_0(1-p_c)\ln(1/\lambda)}\right\}^{1/2} \tag{101}$$

in three dimensions. The corresponding asymptotic forms of $\langle R^2(t)\rangle$ and $\psi(t)$ as $t\to\infty$ are

$$\langle R^2(t)\rangle \propto t^{1/2}\,, \quad \psi(t)\propto t^{-3/2} \tag{102}$$

in two dimensions, and

$$\langle R^2(t)\rangle \propto t^{1/2}(\ln t)^{-1/2}\,, \quad \psi(t)\propto t^{-3/2}(\ln t)^{-1/2} \tag{103}$$

in three dimensions. More strongly nonuniversal behavior is found if we take $h(w)\sim cw^{-\alpha}$, as $w\to 0^+$, $0<\alpha<1$.

In this case, it can be shown from Eq. (88) that

$$c W_m(\lambda)^2 \{(1-p_c) W_m(\lambda)/p_c\}^{-\alpha} \frac{\pi}{\sin(\pi\alpha)} \sim \frac{\lambda p_c \mathcal{S}(\lambda/W_m(\lambda))}{1-p_c} , \tag{104}$$

and so

$$W_m(\lambda) \sim \left\{\frac{(1-\alpha)A \sin(\pi\alpha)}{(2-\alpha)4\pi^2 c}\right\}^{1/(2-\alpha)} \times \left\{\frac{p_c}{1-p_c}\right\}^{(1-\alpha)/(2-\alpha)} (\lambda \ln 1/\lambda)^{1/(2-\alpha)} \tag{105}$$

in two dimensions, and

$$W_m(\lambda) \sim \left\{\frac{\mathcal{S}(0) \sin(\pi\alpha)}{\pi c}\right\}^{1/(2-\alpha)} \left\{\frac{p_c}{1-p_c}\right\}^{(1-\alpha)/(2-\alpha)} \lambda^{1/(2-\alpha)} \tag{106}$$

in three dimensions. The mean-squared displacements at long times are given by

$$\langle R^2(t)\rangle \propto t^{(1-\alpha)/(2-\alpha)} (\ln t)^{1/(2-\alpha)} \quad \text{in two dimensions} \tag{107}$$

and

$$\langle R^2(t)\rangle \propto t^{(1-\alpha)/(2-\alpha)} \quad \text{in three dimensions} . \tag{108}$$

C. The binary distribution

We now examine the binary distribution (2) (for which $h_{-1} = 1/w_0$) in more detail. Equation (75) reduces to

$$W_m(\lambda)/w_0 = \frac{p - p_c + p_c \epsilon \mathcal{S}(\epsilon)}{1 - p_c + p_c \epsilon \mathcal{S}(\epsilon)} , \tag{109}$$

and in the dc limit ($\epsilon \to 0$) gives

$$W_m(0)/w_0 = (p - p_c)/(1 - p_c) , \quad \text{if } p \geq p_c , \tag{110}$$

which is Kirkpatrick's result[19] for the EMA theory of dc conductivity of periodic lattices of random resistors.

To estimate the time scale on which diffusive motion is fully developed (i.e., $\langle R^2(t)\rangle/t \simeq$ constant), we analyze the first order correction to $W_m(\lambda)$ as $\lambda \to 0^+$ above the percolation threshold. For two-dimensional lattices we find [using Eq. (11)]

$$W_m(\lambda) \sim \frac{w_0(p-p_c)}{(1-p_c)} \left\{1 + \frac{A p_c (1-p_c)}{4\pi w_0} \left[\frac{1}{(p-p_c)^2} - \frac{1}{(1-p_c)(p-p_c)}\right] \lambda \ln\left[\frac{w_0(p-p_c)}{\lambda(1-p_c)}\right] + \cdots\right\} . \tag{111}$$

Equation (111) is correct (within the EMA) when interpreted as an asymptotic expansion of $W_m(\lambda)$, as $\lambda \to 0^+$, with w_0 and $p - p_c > 0$ held fixed. It is numerically accurate only when

$$\{\lambda/w_0(p-p_c)^2\} \ln\{w_0(p-p_c)/\lambda\} \ll 1 . \tag{112}$$

In three dimensions the asymptotic expansion is

$$W_m(\lambda) \sim \frac{w_0(p-p_c)}{(1-p_c)} \left\{1 + \frac{p_c(1-p_c)}{w_0} \times \left[\frac{1}{(p-p_c)^2} - \frac{1}{(1-p_c)(p-p_c)}\right] \mathcal{S}(0)\lambda + \cdots\right\} . \tag{113}$$

For any fixed values of w_0 and $p - p_c$, the term linear in λ is negligible compared to the constant term when

$$\lambda \ll w_0(p-p_c)^2 . \tag{114}$$

If, in addition to Eq. (110), we also have $p - p_c \ll 1$, we obtain the simpler equation

$$W_m(\lambda) \simeq \frac{w_0(p-p_c)}{(1-p_c)} \left\{1 + \frac{p_c(1-p_c)}{w_0(p-p_c)^2} \mathcal{S}(0)\lambda + \cdots\right\} . \tag{115}$$

A special case of Eq. (115), appropriate for the simple cubic lattice, has been given by Webman.[15] [We correct a minor error in his Eq. (20) for $W_m(\lambda)$ when $p > p_c$ and $\lambda \ll w_0(p-p_c)^2$: replace his $a \equiv 2\,\mathcal{S}(0)$ by $\mathcal{S}(0)$.]

The inequalities (112) and (114) define a natural time scale τ, such that motion is diffusive when $t \gg \tau$, where

$$\{\tau w_0(p-p_c)^2\}^{-1} \ln\{\tau w_0[p-p_c]\} = 1 \tag{116}$$

in two dimensions and

$$w_0 \tau (p-p_c)^2 = 1 \tag{117}$$

in three dimensions. This time scale τ diverges as $p \to p_c^+$. The long-time behavior implied by Eqs. (111) and (113) is

$$\langle R^2(t)\rangle \sim \begin{cases} \dfrac{z w_0(p-p_c)}{(1-p_c)} t\{1 + \mathcal{O}(t^{-1}\ln t)\} & \text{in two dimensions} \quad (118) \\ \dfrac{z w_0(p-p_c)}{(1-p_c)} t\{1 + \mathcal{O}(t^{-1})\} & \text{in three dimensions} . \quad (119) \end{cases}$$

The diffusion constant vanishes linearly with $p - p_c$ {as we established above [Eq. (78)]} for a general percolationlike distribution.

Below the percolation threshold, we note that in one dimension, Eq. (109) can be solved exactly, giving

$$W_m(\lambda) = \frac{\{1-(1-p)^2\}\lambda}{2(1-p)^2 + \lambda/w_0 + 2(1-p)^2\{1 + \lambda/w_0(1-p)^2 + \lambda^2/4w_0^2(1-p)^2\}^{1/2}} = \frac{p(2-p)\lambda}{4(1-p)^2} \left\{1 - \frac{\lambda}{2w_0(1-p)^2} + \mathcal{O}(\lambda^3)\right\} . \tag{120}$$

This result has been given by Odagaki and Lax.[16] In two or more dimensions, we can determine the $\mathcal{O}(\lambda)$ term by writing $\kappa^{-1} = \lim_{\lambda\to 0} W_m(\lambda)/\lambda$ and making the expansion $\mathcal{S}(\epsilon) \sim \mathcal{S}(\kappa) + \mathcal{S}'(\kappa)(\epsilon - \kappa) + \cdots$. We find that

$$W_m(\lambda) = \kappa^{-1}\lambda \left\{1 - \frac{(1-p)\lambda}{\kappa^2 w_0 p_c \{\mathcal{S}(\kappa) + \kappa \mathcal{S}'(\kappa)\}} + \mathcal{O}(\lambda^2)\right\} , \tag{121}$$

where κ is the solution of Eq. (83). In particular, near p_c, we note from Eq. (85) that in three dimensions $\kappa \sim (1 - p/p_c)/\mathcal{S}(0)$, giving

$$W_m(\lambda) \sim \frac{\lambda \mathcal{S}(0) p_c}{p_c - p} \left\{1 - \frac{\mathcal{S}(0) p_c (1-p_c)\lambda}{w_0(p_c-p)^2} + \mathcal{O}(\lambda^2)\right\} . \tag{122}$$

[An analogous formula has been given by Webman[15] for the simple cubic lattice.] The stochastic evolution of the system approaches equilibrium when $t \gg \tau$, with τ defined by Eq. (117).

VI. DISCUSSION

In the present paper, we address the problem of stochastic transport in disordered systems, a problem of wide interest in both the pure and the applied sciences. Our starting point is the discrete linear master equation (1), with the transition rates W_{ij} ($=W_{ji}$) between nearest-neighbor sites randomly distributed, with probability density function $f(w)$, and our goal is the determination of the effective properties of the system, embodied in a characteristic rate or conductivity W_m. A Green function formalism, developed in Sec. II, reduces the master equation to a form suitable for the development of a class of effective medium approximations (EMA's), both for periodic and topologically disordered lattices. We consider here in detail only periodic lattices and develop the simplest EMA (single-bond EMA). In the single-bond EMA, only the statistical distribution of transition rates of one bond of the lattice is taken into account. The remaining bonds of the lattice are assigned the transition rate W_m, and W_m is then determined self-consistently, as discussed in Sec. III.

It is important to realize that although the quantity W_m so determined is independent of spatial position, it must contain some time dependence. To see this, we need only consider the most naive approximate analysis of the master equation, in which all of the transition rates W_{ij} are replaced by a single, constant transition rate $\overline{W}$ [such as the mean rate $\int_0^\infty wf(w)\,dw$]. The resulting equation is a discretization of the diffusion equation for a uniform system and so can predict only diffusive behavior. Consider a distribution of transition rates which is percolationlike, i.e., only a fraction $p<1$ of all bonds of the lattice are active (have $W_{ij}>0$). We know from percolation theory that if p lies below a critical value p_c (the percolation threshold, determined solely by the lattice topology), no connected paths of active bonds span the lattice, and so diffusion or any long-ranged transport is precluded and the approximation fails. When (as in Secs. II and III) the analysis is performed in Laplace transform space, these difficulties are automatically circumvented. The effective transition rate is then a function $W_m(\lambda)$ of the Laplace transform variable λ, and is shown in Sec. IV to correspond in the time domain to a function $\check{W}_m(t)$, which is the memory kernel for a generalized master equation describing the evolution of the system. Given as an initial condition complete localization at a specified lattice site, $W_m(\lambda)$ determines the mean-squared displacement $\langle R^2(t)\rangle$ at subsequent times; it also determines the waiting-time density $\psi(t)$ for an associated continuous-time random walk description of the evolution of the system.

In Sec. V, we give a detailed analysis of the predictions of the single-bond EMA for eight different lattice systems. We pay particular attention to the qualitatively distinct behavior generated by several different classes of transition rate probability density functions $f(w)$. We recover results obtained by other authors using different formalisms[14–16,42] and give a number of extensions, emphasizing the restrictions on $f(w)$ necessary to obtain universal behavior [critical exponents and asymptotic forms independent of the structure of $f(w)$].

Many of our results are summarized in Tables I–IV. Table I lists the eight periodic lattices considered in the present paper, illustrating some effects of variation of dimensionality d and coordination number z. A key mathematical quantity $\mathcal{G}(\epsilon)$ (defined in terms of the Green function of the lattice) determines most of the qualitative features of the stochastic transport, through its asymptotic form as $\epsilon \to 0$. The asymptotic form depends strongly on the dimensionality d of the lattice, but only weakly on the coordination number z. If p denotes the fraction of all bonds which are active [Eq. (56)], the single-bond EMA gives the approximate value $2/z$ for the bond percolation threshold p_c of the lattice, with qualitatively different behavior according as $0<p<p_c$,

TABLE I. Summary of EMA results for topologically ordered lattices, as a function of dimensionality (d), and coordination number (z).

d	Lattice	z	EMA $p_c=2/z$	Exact p_c	$\mathcal{G}(\epsilon)=-G_{ii}(\epsilon)$	
1	linear chain	2	1	1.0000	$\epsilon^{-1/2}(4+\epsilon)^{-1/2}$	
2	hexagonal	3	2/3	0.6527[a]	$A=\sqrt{3}$	$\mathcal{G}(\epsilon)\sim(A/4\pi)\ln(1/\epsilon)$,
	square	4	1/2	0.5000[a]	$A=1$	as $\epsilon\to 0^+$
	triangular	6	1/3	0.3473[a]	$A=1/\sqrt{3}$	
3	diamond	4	1/2	0.388[b]	$\mathcal{G}(0)=0.448\,22$[e]	
	simple cubic	6	1/3	0.2495[c]	$\mathcal{G}(0)=0.252\,73$[f]	$\mathcal{G}(\epsilon)\to\mathcal{G}(0)<\infty$,
	body-centered cubic	8	1/4	0.178[d]	$\mathcal{G}(0)=0.174\,15$[f]	as $\epsilon\to 0^+$
	face-centered cubic	12	1/6	0.119[d]	$\mathcal{G}(0)=0.112\,06$[f]	

[a]Exact values (Ref. 67); hexagonal $1-2\sin(\pi/18)$, square 1/2, triangular $2\sin(\pi/18)$.
[b]Numerical estimate (Ref. 68).
[c]Numerical estimate (Ref. 69).
[d]Numerical estimate (Ref. 70).
[e]From Ref. 58.
[f]From Eqs. (A25)–(A27).

TABLE II. EMA prediction of behavior of $\lim_{t\to\infty}\langle R^2(t)\rangle$, as $p\to p_c^-$. (The one-dimensional result is correct for $0<p<p_c=1$; the two- and three-dimensional results are the leading terms of asymptotic expansions.)

d	1	2	3
$\lim_{t\to\infty}\langle R^2(t)\rangle$	$\frac{p(2-p)}{2(1-p)^2}$	$\frac{zA\ln\{(1-p/p_c)^{-1}\}}{4\pi(1-p/p_c)}$	$\frac{z\mathcal{G}(0)}{(1-p/p_c)}$

TABLE IV. EMA prediction of ultimate growth with time of $\langle R^2(t)\rangle$ for $p=p_c$ (all multiplicative constants have been omitted).

d	$h_{-1}<\infty$	$h(w)\to h_0$ as $w\to 0$	$h(w)\sim cw^{-\alpha}$ as $w\to 0$ $(0<\alpha<1)$
1	t	$t/\ln t$	$t^{(2-2\alpha)/(2-\alpha)}$
2	$t^{1/2}(\ln t)^{1/2}$	$t^{1/2}$	$t^{(1-\alpha)/(2-\alpha)}(\ln t)^{1/(2-\alpha)}$
3	$t^{1/2}$	$t^{1/2}(\ln t)^{-1/2}$	$t^{(1-\alpha)/(2-\alpha)}$

$p=p_c$, or $p_c<p\leq 1$. One measure of the accuracy of the single-bond EMA is the degree of precision with which known values of p_c are reproduced. In one and two dimensions, exact values of p_c are known, and we see from Table I that the EMA returns the exact value of p_c for the linear chain and square lattice and quite good approximations for the hexagonal and triangular lattices. The discrepancy is larger in three dimensions,[71] where numerical estimates of the correct value of p_c are available, and p_c is well approximated by $1.5/z$.[43] However, the single-bond EMA correctly predicts that the value of p_c decreases as the coordination number increases. These observations, together with evidence[44,45] that the critical exponent t_c for dc conductivity [conductivity $\propto(p-p_c)^{t_c}$, as $p\to p_c^+$] is better approximated by effective medium theories in two dimensions than in three,[72] suggests that the single-bond EMA of the present paper should follow the exact result more closely in two dimensions than in three. Some exact analysis for the one-dimensional problem[41,46,47] confirms the excellence of the EMA in one dimension: the long-time behavior of the mean-squared displacement is exactly reproduced.[48]

We demonstrate in Sec. V that in two or more dimensions, the EMA predicts ultimately diffusive behavior when all bonds are active (have nonzero transition rate). To obtain qualitatively different behavior, it is necessary to turn to one-dimensional lattices, or to percolationlike distributions with only a fraction $p<1$ of all bonds active $f(w)=(1-p)\,\delta_+(w)+ph(w)$. In the latter case, with $\langle R^2(t)\rangle$ the mean-squared displacement at time t, we find in two or three dimensions that as $t\to\infty$

(i) $\langle R^2(t)\rangle\to$ constant, if $p<p_c$ (localization),

(ii) $\langle R^2(t)\rangle\to\infty$, but $\langle R^2(t)\rangle/t\to 0$, if $p=p_c$ (fractal transport),

(iii) $\langle R^2(t)\rangle\propto t$, if $p>p_c$ (diffusion).

The existence of both diffusive and nondiffusive behavior in one dimension when $p=1$ [depending on the structure of $f(w)$] arises from the coincidence of the percolation threshold (where for $d\geq 2$ there is no diffusion) with unity (where for $d\geq 2$ there is diffusion).

Table II shows the behavior of the equilibrium mean-squared displacement ($\lim_{t\to\infty}\langle R^2(t)\rangle$) as $p\to p_c^-$. This behavior is, to leading order, independent of the structure of the probability density function $h(w)$ for the transition rates of active bonds. It reflects the growth in mean size of clusters (subsets of the lattice connected by active bonds) as the percolation threshold is approached. The clusters grow more rapidly in low dimensions, for reasons similar to those underlying Pólya's theorem (Sec. III) on the change with dimension of the qualitative behavior of random walks: it is easier for growing clusters to find each other in a low dimensional space.

Above the percolation threshold ($p>p_c$, $d=2$ or $d=3$), the EMA predicts diffusion at large times, $\langle R^2(t)\rangle\sim 2dDt$, with the diffusion constant D vanishing as $p\to p_c^+$. We give the EMA predictions of D as $p\to p_c^+$ in Table III. If the distribution $h(w)$ of transition rates of active bonds satisfies the moment condition

$$h_{-1}=\int_0^\infty \frac{h(w)\,dw}{w}<\infty\,, \tag{123}$$

i.e., bonds of very low activity are sufficiently rare, we have universal behavior [critical exponents independent of $h(w)$]. However, if $h_{-1}=\infty$, the critical exponents are distribution dependent. A detailed analysis of the binary distribution (2) reveals the existence of time scales defined by Eqs. (116) and (117) which must be exceeded before diffusive behavior is observable.

Table IV summarizes the large time behavior of $\langle R^2(t)\rangle$, the mean-squared displacement, for lattices at the percolation threshold. We again encounter universality if $h_{-1}<\infty$, and nonuniversality otherwise. The waiting-time distribution $\psi(t)$ for the associated continuous-time random walk is longtailed, so that the mean time $\langle t\rangle$ between steps is infinite.

The extension of the basic ideas of the present paper to topologically disordered lattices and to the construction of higher order effective medium approximations will be considered elsewhere.[21,22] The basic formalism admits certain other extensions, to which we can only allude briefly here. The point of departure of the basic analysis is the Laplace transformed version (4) of the linear

TABLE III. EMA prediction of asymptotic form of diffusion constant D as $p\to p_c^+$.

d	$h_{-1}<\infty$	$h(w)\to h_0$ as $w\to 0$	$h(w)\sim cw^{-\alpha}$, as $w\to 0$ $(0<\alpha<1)$
2, 3	$\frac{z(p-p_c)}{2d\,h_{-1}(1-p_c)}$	$\frac{z(p-p_c)}{2d\,h_0(1-p_c)\ln\{[p-p_c]^{-1}\}}$	$\frac{z}{2d}\left\{\frac{\sin(\pi\alpha)}{c\pi p_c^\alpha}\right\}^{1/(1-\alpha)}\frac{(p-p_c)^{\alpha/(1-\alpha)}}{(1-p_c)}$

master equation (1), so that any stochastic process in a disordered system which can be mapped onto Eq. (4) can be analyzed in a similar fashion. One may consider a generalized master equation

$$\frac{dP_i}{dt} = \sum_{j\in\{i\}} \int_0^t d\tau\, \check{W}_{ij}(t-\tau)\{P_j(\tau) - P_i(\tau)\}\,, \tag{124}$$

with the memory kernels $\check{W}_{ij}(t)$ functions of time parametrized by one or more random variables, e.g., $\check{W}_{ij}(t) = \alpha e^{-\beta t}$, with α and β assigned probability distributions based on some microscopic model. (Here $\{i\}$ denotes the set of nearest neighbors of site i.) In Laplace transform space we obtain

$$\lambda\hat{P}_i(\lambda) - \delta_{i0} = \sum_{j\in\{i\}} \hat{W}_{ij}(\lambda)\{\hat{P}_j(\lambda) - \hat{P}_i(\lambda)\}\,, \tag{125}$$

an equation to which we may apply the analysis of the present paper, and which should yield an even wider class of possible stochastic behaviors.

Finally,[73] we note that the restriction of interactions to nearest-neighbor sites is unnecessary, although the Green functions become more complicated. Since random walks with infinite mean-squared displacement per step yield a variety of nondiffusive behaviors,[50] which may be further enriched by the introduction of long-tailed waiting-time distributions[51] an extension of the EMA to continuous-time random walks or generalized master equations with long-ranged transitions could yield very interesting results.

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APPENDIX

We indicate briefly how a Green function G_{ik}, defined by Eq. (8), can be found for a variety of periodic lattices. Equations equivalent to Eq. (8), arise in numerical analysis,[54] in the theory of lattice dynamics within the harmonic approximation[55] and in the theory of lattice random walks.[27] Although they have been extensively studied in these contexts (so that we can refer the reader to the literature for most of the details), it is useful to collect here all of the results of interest using a consistent notation.

Consider first a simple cubic lattice in d dimensions (so that each site has coordination number $z = 2d$). Since G_{ik} depends only on the relative positions of sites i and k, there is no loss of generality in writing $k = (0, 0, \ldots, 0)$ and $i = (m_1, m_2, \ldots, m_d)$ (with $m_1, m_2, \ldots m_d$ integers) and defining $G(m_1, m_2, \ldots, m_d) \equiv G_{ik}$. Equation (8) reduces to the difference equation

$$\begin{aligned}(2d+\epsilon)G(m_1, m_2, \ldots, m_d) - \{&G(m_1+1, m_2, \ldots, m_d) \\ &+ G(m_1-1, m_2, \ldots, m_d) + G(m_1, m_2+1, \ldots, m_d) \\ &+ G(m_1, m_2-1, \ldots, m_d) + \cdots + G(m_1, m_2, \ldots, m_d+1) \\ &+ G(m_1, m_2, \ldots, m_d-1)\} = -\delta_{m_1 0}\delta_{m_2 0}\cdots\delta_{m_d 0}\,,\end{aligned} \tag{A1}$$

which can be solved by use of a discrete Fourier transform. We define

$$\begin{aligned}\tilde{G}(\theta_1, \theta_2, \ldots, \theta_d) = \sum_{m_1=-\infty}^{\infty}\sum_{m_2=-\infty}^{\infty}\cdots\sum_{m_d=-\infty}^{\infty} &\exp(im_1\theta_1 + im_2\theta_2 \\ &+ \cdots + im_d\theta_d) G(m_1, m_2, \ldots, m_d)\,,\end{aligned} \tag{A2}$$

and find that

$$\{2d + \epsilon - 2(\cos\theta_1 + \cos\theta_2 + \cdots + \cos\theta_d)\}\tilde{G}(\theta_1, \theta_2, \ldots, \theta_d) = -1\,. \tag{A3}$$

Inverting the discrete Fourier transform gives

$$G(m_1, m_2, \ldots, m_d) = -\frac{1}{2}\frac{1}{(2\pi)^d}\int\!\!\int_{-\pi}^{\pi}\cdots\int \frac{d\theta_1 d\theta_2\cdots d\theta_d \exp(-im_1\theta_1 - im_2\theta_2 - \cdots - im_d\theta_d)}{d + \frac{\epsilon}{2} - \cos\theta_1 - \cos\theta_2 - \cdots - \cos\theta_d} \tag{A4}$$

$$= -\frac{1}{2}\frac{1}{(2\pi)^d}\int\!\!\int_{-\pi}^{\pi}\cdots\int d\theta_1 d\theta_2\cdots d\theta_d \exp(-im_1\theta_1 - im_2\theta_2 - \cdots - im_d\theta_d)$$

$$\cdot \int_0^\infty dt \exp[-(d+\tfrac{1}{2}\epsilon)t + (\cos\theta_1 + \cos\theta_2 + \cdots + \cos\theta_d)t] = -\frac{1}{2}\int_0^\infty dt \exp[-(d+\tfrac{1}{2}\epsilon)t] I_{m_1}(t) I_{m_2}(t)\cdots I_{m_d}(t)\,, \tag{A5}$$

where we have used the integral representation of the modified Bessel function

$$I_m(t) = \frac{1}{2\pi}\int_{-\pi}^{\pi}\cos(m\theta)\exp(t\cos\theta)d\theta\,. \tag{A6}$$

The necessity of keeping $\mathrm{Re}(\epsilon) \geq 0$, and of insisting that $\epsilon \neq 0$ when $d \leq 2$ is apparent from the asymptotic expansion

$$I_m(t) = (2\pi t)^{-1/2} e^t\{1 + \mathcal{O}(t^{-1})\}\,, \quad \text{as } t \to \infty\,. \tag{A7}$$

In the case $d = 1$ (a linear chain) the Green function can be evaluated in terms of elementary functions, by means of a known Laplace transform pair[56]:

$$G(m) = -\epsilon^{-1/2}(4+\epsilon)^{-1/2}\{1 + \tfrac{1}{2}\epsilon - \epsilon^{1/2}(1+\tfrac{1}{4}\epsilon)^{1/2}\}^{|m|}\,. \tag{A8}$$

There is an inverse square root singularity as $\epsilon \to 0^+$. To obtain an expression for the singular behavior as $\epsilon \to 0^+$ of the Green function for the square lattice ($d = 2$) we note that for any $\epsilon > 0$,

$$\begin{aligned}G_{sq}(m_1, m_2) = &-\frac{1}{2}\int_0^1 dt \exp(-2t - \tfrac{1}{2}\epsilon t) I_{m_1}(t) I_{m_2}(t) \\ &-\frac{1}{2}\int_1^\infty dt \exp(-2t - \tfrac{1}{2}\epsilon t)\{I_{m_1}(t) I_{m_2}(t) - (2\pi t)^{-1} e^{2t}\} \\ &-\frac{1}{4\pi}\int_1^\infty \frac{dt}{t}\exp(-\tfrac{1}{2}\epsilon t).\end{aligned} \tag{A9}$$

Only the third term on the right-hand side diverges as $\epsilon \to 0^+$ and its behavior as $\epsilon \to 0^+$ may be established by recognizing it as the exponential integral[57]

$$E_1(z) = \int_z^\infty \frac{dt}{t} \exp(-t) \sim -\ln z, \quad \text{as } z \to 0 . \tag{A10}$$

Thus, for the square lattice

$$G_{sq}(m_1, m_2) \sim -(4\pi)^{-1} \ln(1/\epsilon), \quad \text{as } \epsilon \to 0^+ . \tag{A11}$$

Integral representations for the Green functions of other periodic lattices can be obtained by mapping the sites of these lattices onto a subset of the sites of a simple cubic lattice, and solving the equivalent problem of a simple cubic lattice with some or all of the nearest-neighbor couplings removed, and some couplings to more remote sites added.[27] We consider first two three-dimensional lattices: the body-centered cubic (BCC) lattice, with coordination number $z = 8$, and the face-centered cubic (FCC) lattice, with coordination number $z = 12$. As before, we write $k = (0, 0, 0)$, $i = (m_1, m_2, m_3)$, and $G_{ik} = G(m_1, m_2, m_3)$. Any site j of the BCC lattice is coupled to eight sites, whose coordinates differ from those of j by $(\pm 1, \pm 1, \pm 1)$, while any site j of the FCC lattice is coupled to 12 sites whose coordinates differ from those of j by $(\pm 1, \pm 1, 0)$, $(\pm 1, 0, \pm 1)$, or $(0, \pm 1, \pm 1)$. Writing down the appropriate difference equations for $G(m_1, m_2, m_3)$ and employing discrete Fourier transforms as above, we obtain

$$G_{BCC}(m_1, m_2, m_3) = -\frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} \frac{d\theta_1 d\theta_2 d\theta_3 \exp(-im_1\theta_1 - im_2\theta_2 - im\theta_3)}{8 + \epsilon - 8\cos\theta_1 \cos\theta_2 \cos\theta_3} , \tag{A12}$$

$$G_{FCC}(m_1, m_2, m_3) = -\frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} \frac{d\theta_1 d\theta_2 d\theta_3 \exp(-im_1\theta_1 - im_2\theta_2 - im_3\theta_3)}{12 + \epsilon - 4(\cos\theta_1 \cos\theta_2 + \cos\theta_2 \cos\theta_3 + \cos\theta_3 \cos\theta_1)} . \tag{A13}$$

[These Green functions take the value 0 at those sites (m_1, m_2, m_3) of the underlying simple cubic lattice which are not sites of the BCC or FCC lattice.] For other three-dimensional lattices the analysis is not so straightforward, though Ishioka and Koiwa[58] have found the Green function for the diamond lattice ($z = 4$).

The triangular lattice ($z = 6$, $d = 2$) may be analyzed as a sheared square lattice with one diagonal interaction[59] or, more simply, as a square lattice with site j coupled to the six sites whose coordinates differ from those of j by $(\pm 2, 0)$ or $(\pm 1, \pm 1)$, as shown in Fig. 1. Proceeding as above, one may show[60] that

$$G_{tri}(m_1, m_2) = -\frac{1}{(2\pi)^2} \iint_{-\pi}^{\pi} \frac{d\theta_1 d\theta_2 \exp(-im_1\theta_1 - im_2\theta_2)}{6 + \epsilon - 4\cos\theta_1 \cos\theta_2 - 2\cos 2\theta_1} , \tag{A14}$$

with this integral vanishing identically for values of (m_1, m_2) which do not correspond to sites of the triangular lattice. At all other sites, when $\epsilon = 0$ the integrand has nonintegrable singularities at the central point $(\theta_1, \theta_2) = (0, 0)$, and also at the four corners $(\theta_1, \theta_2) = (\pm\pi, \pm\pi)$. The leading order behavior of $G_{tri}(m_1, m_2)$ as $\epsilon \to 0^+$ can be obtained by analyzing the behavior of the integrand within a small ellipsoid centered on each of these singular points. Near (0, 0), the integrand is, to leading order, $(\epsilon + 2\theta_1^2 + 6\theta_2^2)^{-1}$, and for any $\delta > 0$

$$-\frac{1}{(2\pi)^2} \iint_{6\theta_1^2 + 2\theta_2^2 \le \delta^2} \frac{d\theta_1 d\theta_2}{\epsilon + 6\theta_1^2 + 2\theta_2^2} = -\frac{1}{8\sqrt{3}\pi^2} \iint_{x^2+y^2 \le \delta^2} \frac{dx\, dy}{\epsilon + x^2 + y^2}$$
$$= -\frac{1}{4\sqrt{3}\,\pi} \int_0^\delta \frac{dr \cdot r}{\epsilon + r^2} \sim -\frac{1}{8\sqrt{3}\,\pi} \ln(1/\epsilon) , \quad \text{as } \epsilon \to 0^+ . \tag{A15}$$

Each of the corner points $(\pm\pi, \pm\pi)$ gives a similar contribution, weighted by a factor of 1/4, and we deduce that

$$G_{tri}(m_1, m_2) \sim -\frac{1}{4\sqrt{3}\,\pi} \ln(1/\epsilon) , \quad \text{as } \epsilon \to 0^+ . \tag{A16}$$

The hexagonal, or "honeycomb" lattice ($z = 3$, $d = 2$), is more difficult to analyze, because it is not translationally invariant. Horiguchi[60] has shown how its Green function may be calculated in terms of the Green function for the triangular lattice, but we offer here a briefer and more direct derivation. We map the hexagonal lattice onto a square lattice as shown in Fig. 2, with the inhomogeneous term in the equation defining G_{ik} located at (0, 0). To accommodate the two distinct types of sites shown in Fig. 3, we introduce auxiliary Green functions

$$G_{hex}(2m_1, m_2) = \phi(m_1, m_2) ,$$
$$G_{hex}(2m_1 + 1, m_2) = \psi(m_1, m_2) , \tag{A17}$$

and reduce Eq. (8) to a pair of simultaneous difference equations

$$(3 + \epsilon)\,\phi(m_1, m_2) - \{\psi(m_1, m_2) + \psi(m_1 - 1, m_2 - 1) + \psi(m_1 - 1, m_2 + 1)\} = -\delta_{m_1 0}\delta_{m_2 0} , \tag{A18}$$

$$(3 + \epsilon)\,\psi(m_1, m_2) - \{\phi(m_1, m_2) + \phi(m_1 + 1, m_2 + 1) + \phi(m_1 + 1, m_2 - 1)\} = 0 . \tag{A19}$$

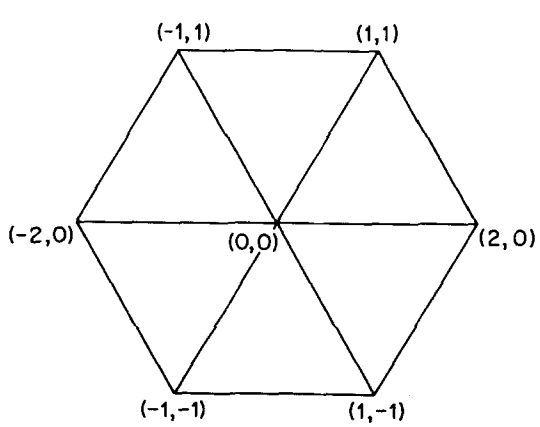

FIG. 1. The sites of the triangular lattice can be mapped onto a subset of the sites of the square lattice. We show here the coordinates assigned to the origin of the triangular lattice and its six nearest neighbors.

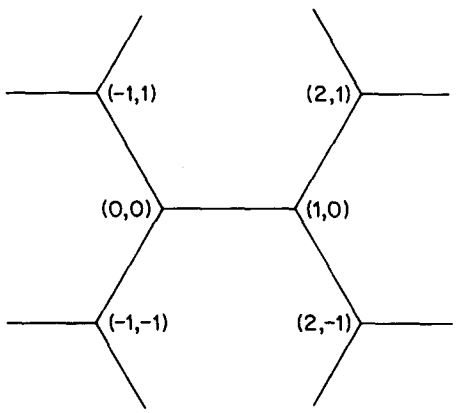

FIG. 2. A mapping of the sites of the hexagonal (honeycomb) lattice onto a subset of the sites of the square lattice. We show here the coordinates assigned to six of the hexagonal lattice sites.

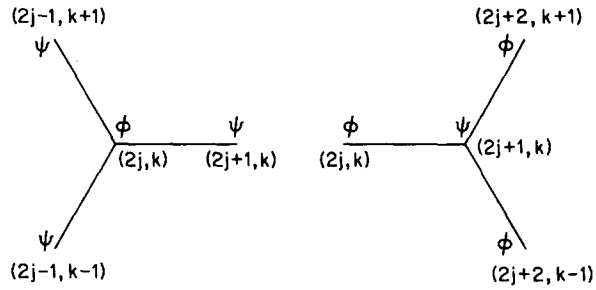

FIG. 3. The two possible configurations of any site of the hexagonal lattice: a ϕ site, surrounded by three ψ sites, and a ψ site, surrounded by three ϕ sites.

These equations may be converted to a pair of simultaneous algebraic equations by taking a discrete Fourier transform. Solution of these equations and inversion of the transform gives

$$G_{\rm hex}(2m_1, m_2) = \frac{-1}{(2\pi)^2} \times \iint_{-\pi}^{\pi} \frac{d\theta_1 d\theta_2 \exp(-im_1\theta_1 - im_2\theta_2)(3+\epsilon)}{(3+\epsilon)^2 - 1 - 4\cos\theta_1\cos\theta_2 - 4\cos^2\theta_2} , \tag{A20}$$

$$G_{\rm hex}(2m_1+1, m_2) = \frac{-1}{(2\pi)^2} \times \iint_{-\pi}^{\pi} \frac{d\theta_1 d\theta_2 \exp(-im_1\theta_1 - im_2\theta_2)(1+2\exp(-i\theta_1)\cos\theta_2)}{(3+\epsilon)^2 - 1 - 4\cos\theta_1\cos\theta_2 - 4\cos^2\theta_2} . \tag{A21}$$

[It can again be shown that the Green function defined by Eqs. (A20) and (A21) vanishes, as it should, at those points of the square lattice which do not correspond to sites of the hexagonal lattice.] The behavior of the Green function as $\epsilon \to 0^+$ can be analyzed as for the triangular lattice. We find

$$G_{\rm hex}(m_1, m_2) \sim -\frac{\sqrt{3}}{4\pi} \ln(1/\epsilon) . \tag{A22}$$

For the purposes of the present paper, we require only the values of the Green function for each lattice at the origin $[(m_1, m_2, \ldots, m_d) = (0, 0, \ldots, 0)]$ and at each of its nearest neighbors. For any lattice with the property that all sites are topologically equivalent (a class which includes all those discussed in this appendix), symmetry considerations guarantee that the Green function takes the same value at each of the nearest neighbors of the origin. Setting $i = k$ in Eq. (8), we then have

$$(z+\epsilon)\,G(\text{origin}) - zG(\text{nearest neighbor}) = -1 . \tag{A23}$$

We need, therefore, to evaluate the Green function only at the origin.

It is possible to reduce the integral representations of the Green function at the origin for each of the lattices considered in this appendix to complete elliptic integrals (for $d = 2$) or products of complete elliptic integrals (for $d = 3$). The analysis of the three-dimensional cases is extremely complicated[61] and we shall not give the formulas here, except for the special case $\epsilon = 0$, for which Watson[62] has derived relatively compact expressions in terms of the gamma function and the complete elliptic integral of the first kind[63]

$$\mathrm{K}(k) = \int_0^{\pi/2} \{1 - k^2\sin^2\phi\}^{-1/2} d\phi . \tag{A24}$$

Watson's results are

$$I_{\rm BCC} \equiv \frac{1}{\pi^3}\iiint_0^{\pi} \frac{du\,dv\,dw}{1-\cos u\cos v\cos w} = \frac{1}{4\pi^3}\Gamma(\tfrac{1}{4})^4 \simeq 1.3932 , \tag{A25}$$

$$I_{\rm FCC} \equiv \frac{1}{\pi^3}\iiint_0^{\pi} \frac{du\,dv\,dw}{3-\cos u\cos v-\cos v\cos w-\cos w\cos u} = \frac{3}{2^{14/3}\pi^4}\Gamma(\tfrac{1}{3})^6 \simeq 0.4482 , \tag{A26}$$

$$I_{\rm SC} \equiv \frac{1}{\pi^3}\iiint_0^{\pi} \frac{du\,dv\,dw}{3-\cos u-\cos v-\cos w} = (18+12\sqrt{2}-10\sqrt{3}-7\sqrt{6})(2K_2/\pi)^2 \simeq 0.505\,46 , \tag{A27}$$

where

$$K_2 = \mathrm{K}([2-\sqrt{3}][\sqrt{3}-\sqrt{2}]) . \tag{A28}$$

It has recently been established (Glasser, personal communication, 1982) that

$$I_{\rm SC} = \frac{\sqrt{6}}{96\pi^3}\Gamma(\tfrac{1}{24})\Gamma(\tfrac{5}{24})\Gamma(\tfrac{7}{24})\Gamma(\tfrac{11}{24}) , \tag{A29}$$

an earlier expression of Glasser and Zucker[64] for $I_{\rm SC}$ is in error.

It is relatively easy to evaluate the Green function at the origin for the two-dimensional lattices and we simply quote the results here[59,66]

$$\text{Square:}\quad G(0,0) = -\frac{1}{2\pi}\left(1+\frac{\epsilon}{4}\right)^{-1}\mathrm{K}\left(\left[1+\frac{\epsilon}{4}\right]^{-1}\right) , \tag{A30}$$

Triangular: $$G(0,0) = -\frac{2}{\pi}\{(9+\epsilon)^{1/2}-1\}^{-3/2} \times\{(9+\epsilon)^{1/2}+3\}^{-1/2} K\left(\frac{4(9+\epsilon)^{1/4}}{\{(9+\epsilon)^{1/2}-1\}^{3/2}\{(9+\epsilon)^{1/2}+3\}^{1/2}}\right), \quad \text{(A31)}$$

Hexagonal: $$G(0,0) = -\frac{2(3+\epsilon)}{\pi(2+\epsilon)^{3/2}(6+\epsilon)^{1/2}} \times K\left(\frac{4(\epsilon+3)^{1/2}}{(\epsilon+2)^{3/2}(\epsilon+6)^{1/2}}\right). \quad \text{(A32)}$$

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