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Energy-projected effective-medium theory of long-range hopping on energetically disordered lattices

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We introduce energy-projected equations of motion to treat the diffusive transport of charge carriers that undergo long-range (i.e., greater than nearest-neighbor) hopping among the sites of an energetically disordered lattice. This approach leads naturally to an energy-projected effective-medium theory for treating such systems. Exact expressions for the diffusion constant associated with the energy-projected effective medium theory are obtained. Using the formalism in conjunction with what is normally a rather poor approximation, i.e., the virtual-crystal approximation, we are able to obtain the exact diffusion constant for the long-range symmetric-random-well problem. Effective-medium calculations and numerical simulations are presented for nearest-neighbor and long-range hopping on a disordered binary lattice.

I. INTRODUCTION

In a recent paper¹ we described the application of an energy-projected effective-medium theory (EPEMT) to the problem of nearest-neighbor hopping transport on a binary lattice, i.e., one consisting of sites randomly occupied by ions associated with two different site energies. In this paper we extend the approach of our earlier paper in two ways: (a) to include the possibility of long-range (i.e., greater than nearest-neighbor) hops and (b) to allow an arbitrary distribution of random site energies. Our approach is influenced by previous treatments,^{2–7} which have sought to construct a translationally invariant effective medium that reproduces the configuration-averaged transport coefficients of the actual disordered system of interest. When applied to systems having sites of different energy, approaches of this type suffer a major drawback: all sites in the lattice become equivalent after averaging and the effective-medium jump rate between two sites becomes artificially symmetric with respect to forward and backward jumps. This symmetry, which is artificially imposed by the averaging process, must be broken if detailed information about the contributions to diffusion from different energy states is to be understood. We demonstrate how this average symmetry may be broken through the use of a projected average that takes into account the energy of the states involved in each transition. The rest of the paper is laid out as follows. In Sec. II, we introduce the basic model and the corresponding equations of motion. These are transformed to a set of energy-projected variables, which are then formally averaged to obtain a set of dynamical effective-medium equations. These effective-medium equations are solved in the Appendix to obtain an exact expression for the diffusion constant in terms of effective-medium hopping rates. In Sec. III we discuss applications that demonstrate how very simple approximations to the effective-medium rates can give very good results when used in conjunction with the energy-projection method.

II. DEVELOPMENT OF THE ENERGY-PROJECTED EFFECTIVE MEDIUM

A. Equations of motion

The starting point of our calculation is the long-range hopping transport equation

$$\frac{dP_n}{dt} = \sum_s (W_{n,n+s}P_{n+s} - W_{n+s,n}P_n) \quad (1)$$

appropriate to an energetically disordered solid, in which $P_n(t)$ is the probability at time t of finding the carrier at the site associated with lattice vector $\mathbf{n}=(n_1, \dots, n_d)$. The quantity $W_{m,n} = W(|\mathbf{m}-\mathbf{n}|; \epsilon_m, \epsilon_n)$ denotes the hopping rate from site \mathbf{n} to site \mathbf{m} , which is assumed to depend upon the randomly distributed energies ϵ_m and ϵ_n of the two sites involved in the transition as well as the distance $|\mathbf{m}-\mathbf{n}|$ (measured in lattice spacings) between them. The summation runs over the set $\{s\}$ of vectors connecting all sites in the lattice. Due to the different ionic constituents in the system, the site energies are assumed to be independently distributed random variables drawn from the same distribution $g(\epsilon)$. For simplicity of notation, we assume initially that the distribution of site energies is discrete so that the site energies are drawn from a set $\{\epsilon_\alpha | \alpha=1, \dots, N\}$ corresponding to the different ionic species in the crystal. A given energy ϵ_α occurs at random with a probability x_α equal to the molar concentration of that species, with the sum of the x_α adding up to unity. The hopping rates that appear in the set of equations (1) depend upon the random energies, and so are themselves random variables drawn from a set $\{W_s^{\beta\alpha}\}$, in which (using α and β to distinguish energy labels) $W_s^{\beta\alpha} = W(|s|; \epsilon_\beta, \epsilon_\alpha)$ describes the microscopic rate at which hops proceed to a site of energy ϵ_β separated by a distance s from a site of energy ϵ_α .

At this point we also make the physical assumption that the microscopic hopping rates connecting sites of different energy obey a well-defined detailed balance rela-

tion⁸ that causes the system to approach a unique thermal distribution, independent of its initial condition. If we denote by $f_0(\varepsilon_\alpha)$ the long-time relative site probability to find the carrier at a given site of energy ε_α , then the detailed balance condition ensuring equilibration to this state can be written as follows:

$$W_s^{\beta\alpha} f_0(\varepsilon_\alpha) = W_s^{\alpha\beta} f_0(\varepsilon_\beta). \quad (2)$$

In what follows we do not specify what this distribution is, leaving open the possibility of applying the method to both classical (Boltzmann) and quantum-mechanical (Bose-Einstein or Fermi-Dirac) distributions.

B. Transformation to energy-projected variables

As we have noted, the actual configuration of hopping rates, and thus, the probabilities which appear in (1), depend upon the exact configuration of the lattice. It is convenient to specify a particular configuration by means of a complete set $\{\zeta_n^\alpha\}$ of indicator variables defined so that

$$\zeta_n^\alpha \equiv \begin{cases} 1 & \text{if site } n \text{ is of type } \alpha, \\ 0 & \text{otherwise.} \end{cases} \quad (3)$$

These variables can also be used to define a projection $P_n^\alpha(t) \equiv P_n(t) \zeta_n^\alpha$ of the probability $P_n(t)$ onto those configurations having an ion of type α at site \mathbf{n} . (Alternatively, the quantity P_n^α represents the joint probability that the carrier is at site \mathbf{n} and that site \mathbf{n} is of species α .) Through straightforward manipulations, we obtain an exact rewriting of the equations of motion (1) in terms of these new variables. After performing a Laplace transform over time,

$$\tilde{P}_n^\alpha(z) \equiv \int_0^\infty dt P_n^\alpha(t) \exp(-zt), \quad (4)$$

in which $\tilde{P}_n^\alpha(z)$ denotes the Laplace transform at frequency z of the probability $P_n^\alpha(t)$, we obtain

$$z\tilde{P}_n^\alpha(z) - P_n^\alpha(0) = \sum_{s,\beta} W_{n,n+s}^{\alpha\beta} \tilde{P}_{n+s}^\beta(z) - W_{n+s,n}^{\beta\alpha} \tilde{P}_n^\alpha(z), \quad (5)$$

in which we have introduced $W_{n,n+s}^{\alpha\beta} \equiv W_s^{\alpha\beta} \zeta_n^\alpha$. It is to be emphasized that in obtaining these projected equations, which formally resemble the equations for a system with multiple states per site, no approximations have been made. Note that by giving the site probabilities an energy index, we have formally increased the number of variables in the problem by a factor of N , the number of ionic species in the crystal. However, in any particular configuration many of the projected probabilities are identically equal to zero. This increased complexity has the advantage that when the solutions to these equations are averaged, a structure is obtained which retains the distinction between different kinds of sites.

C. Effective-medium equations

Configuration averaging over the disorder will lead to a *translationally invariant* set of equations,

$$z\tilde{P}_n^\alpha(z) - P_n^\alpha(0) = \sum_{s,\beta} [\omega_s^{\alpha\beta}(z) p_{n+s}^\beta(z) - \omega_s^{\beta\alpha}(z) p_n^\alpha(z)], \quad (6)$$

for the configuration-averaged (but now projected) probabilities $p_n^\alpha(z) \equiv \langle \tilde{P}_n^\alpha(z) \rangle$. We will refer to the $p_n^\alpha(z)$ as effective-medium probabilities, and take the point of view that these (Laplace-transformed) equations of motion define the effective medium that we seek. These new equations now evolve under the action of (frequency-dependent) effective-medium hopping rates $[\omega_s^{\alpha\beta}(z)]$ which connect sites of each type on the energy-projected lattice. Note that Eq. (6) is equivalent to a nonlocal integro-differential equation (or generalized master equation) in the time domain governed by memory kernels $\omega_s^{\beta\alpha}(t)$. This form for the averaged equations has been shown⁹ under rather general conditions to be an exact consequence of the averaging process (which can be viewed as a linear projection) and of the linearity of the original nonaveraged master equation. Thus, although at this point we have not specified exactly what the effective medium rates are, we have made no actual approximations.

The problem reduces at this point to (a) the determination of the effective-medium hopping rates $\omega_s^{\alpha\beta}(z)$, and (b) the calculation of the diffusion constant as a function of these effective-medium rates. The first step, typically, is the hard part of the problem and is, in most theories, the point to which approximation schemes are usually addressed. In what follows we demonstrate that better (sometimes exact) results can be obtained using lower order approximations on these new projected equations than would be obtained without projection. In spite of the complexity introduced into the equations by the energy projection, the second step (calculating the diffusion constant D as a function of the effective-medium rates) can be done exactly. The details of this calculation are presented in the Appendix, where we derive the following simple expression:

$$D = \sum_\alpha D_\alpha \rho_\alpha, \quad (7)$$

for the effective-medium diffusion constant, where

$$D_\alpha = \frac{1}{2d} \sum_{\beta,s} \omega_s^{\beta\alpha} s^2. \quad (8)$$

In these expressions, we have dropped the dependence on z in referring to the zero-frequency limit, so that $\omega_s^{\alpha\beta} \equiv \omega_s^{\alpha\beta}(0)$. Thus, the diffusion constant depends only on the steady-state ($z=0$) limit of the rates $\omega_s^{\alpha\beta}(z)$, as might be expected. The quantity ρ_α appearing in (7) represents the steady-state or equilibrium population, normalized to one particle, for all sites of energy ε_α , i.e.,

$$\rho_\alpha \equiv \lim_{t \rightarrow \infty} \sum_n \langle P_n^\alpha(t) \rangle = Z^{-1} x_\alpha f_0(\varepsilon_\alpha), \quad (9)$$

where $Z = \sum_\alpha x_\alpha f_0(\varepsilon_\alpha)$. The right-hand side of (9) follows from the assumption of a unique equilibrium distribution, and shows that the ρ_α 's are, in principle, calculable *a priori* from a knowledge of the concentrations and site energies of the constituent ions. From the diffusion

constant, through Einstein's relation, one can then obtain the conductivity. Thus, once the hopping rates $\omega_s^{\alpha\beta}$ appearing in the energy-projected effective-medium equations are specified, the asymptotic transport properties follow directly from Eqs. (7)–(9).

III. APPLICATIONS

A. Symmetric well model—exact results

We now discuss the more difficult problem of evaluating and/or approximating the effective medium rates $\omega_s^{\alpha\beta}$. Treatments which do not employ energy projection often proceed by defining a reference system about which to expand, and performing perturbation theory on the Green's function, treating the deviation of the actual system as a perturbation. This leads to various decoupling schemes,³ which vary in complexity from the simple virtual-crystal approximation (VCA) to the self-consistent coherent-potential approximation (CPA). In the VCA, the effective-medium hopping rates are approximated by the configuration-averaged values of the rates that actually occur in the disordered system. The resulting description is often qualitatively inaccurate. In the CPA, the effective medium itself is formally chosen as the reference system, the effect of fluctuations about the effective medium being required to vanish upon averaging.

In the energy-projected effective-medium approximation, excellent results can often be obtained using the simplest approximation (i.e., the VCA) to the energy-projected equations. Consider, for example, the symmetric random well model^{2–4} (sometimes called the random trapping model), in which the hopping rate $W_s^{\alpha\beta} = W_s^\beta$ between two sites is allowed to depend upon the distance between the two sites and upon the energy of the site (or well) from which a particular hop occurs, but is independent of the energy of the site to which the particle is hopping. The physical picture behind this model is that of a set of wells of random depth separated by tall barriers of uniform height. Thus, after obtaining the energy to leave the initial well, the particle is able, in principle, to make jumps to any other site in the crystal with a hopping probability that depends only upon the distance. This is a natural extension of the nearest-neighbor symmetric well problem treated elsewhere,^{2–4} and is in the class of systems to which the projection approach developed above applies. To treat the random well model within the VCA, we let the effective-medium rates $\omega_s^{\alpha\beta}$ appearing in (7) be approximated by the averages of the corresponding rates appearing in the unaveraged Eq. (5). For the random trapping model this entails the approximation $\omega_s^{\alpha\beta} \approx \langle W_{n,n+s}^{\alpha\beta} \rangle = W_s^\beta \langle \xi_n^\alpha \rangle = x_\alpha W_s^\beta$, which when applied to Eqs. (7)–(9) results in the expression

$$2dD = \sum_{s,\beta} s^2 W_s^\beta \rho_\beta, \quad (10)$$

where, with the symmetry of this problem, we are able to explicitly evaluate the populations ρ_β as a function of the hopping rates, i.e.,

$$\rho_\beta = \frac{Z_\beta}{\sum_\alpha Z_\alpha}, \quad (11)$$

in which

$$Z_\beta = \frac{x_\beta}{\sum_s W_s^\beta}. \quad (12)$$

This has previously been shown to be the exact diffusion constant for this model.^{4,10} In this form it is easy to see that the diffusion constant vanishes in the random trapping model for any finite concentration of infinitely deep wells (i.e., traps, for which all exit rates W_s^β vanish). Application of the VCA to the unprojected equations (1), by contrast, leads to very poor results that fail to predict this vanishing of the diffusion constant in the deep trap limit.

B. Long-range hopping on binary disordered lattices

We now consider applications of the EPMT developed in Sec. II to the hopping conductivity of binary lattices, i.e., those possessing two types of sites independently distributed according to the site energy distribution function,

$$\Gamma(\epsilon) = (1-x)\delta(\epsilon - \epsilon_1) + x\delta(\epsilon - \epsilon_2). \quad (13)$$

For convenience, we assume that $\epsilon_1 > \epsilon_2$. In analyzing this system we adopt a similar approach to that taken in Ref. 1, which focused only on the nearest-neighbor case and on the trapping-to-percolation crossover that occurs in that system. The results presented below illustrate the effect that long-range hopping produces on this transition. The basic idea is to use the approximation of the last section (i.e., the VCA),

$$\omega_s^{\alpha\beta}(0) \equiv \langle W_{n+s,n}^{\alpha\beta} \rangle = W_s^{\alpha\beta} \langle \xi_n^\alpha \rangle = x_\alpha W_s^{\alpha\beta} \quad (\alpha \neq \beta) \quad (14)$$

to approximate the effective-medium rates connecting sites of different energy. For states of the same energy, we choose the effective-medium rates $\omega_s^{\alpha\alpha}$ so that they reproduce the conductivity of the isolated sublattice corresponding to sites of energy ϵ_α . In other words, $\omega_s^{\alpha\alpha}$ is chosen so that for each concentration x it reproduces the conductivity of the system when all rates except those between states of the same type α are set equal to zero. In all cases the distance dependence of the rates is kept the same as in the original system. In Ref. 1, nearest-neighbor rates were computed using an analytical approximation which describes diffusion on a site percolating lattice. In this paper we are interested in going beyond the nearest-neighbor case, and so we have used numerically generated data to obtain both the nearest-neighbor and long-range effective-medium rates coupling sites of the same energy. The details of the numerical procedure, which was briefly described in Ref. 1, will appear in a forthcoming publication.^{10,11}

In Fig. 1 we show logarithmic plots of the diffusion constant D , normalized to unity at $x = 1$, as a function of the concentration x of the lower energy sites. In all cases we have taken $W_{11} = W_{22} = W_{21}$, which implies that the

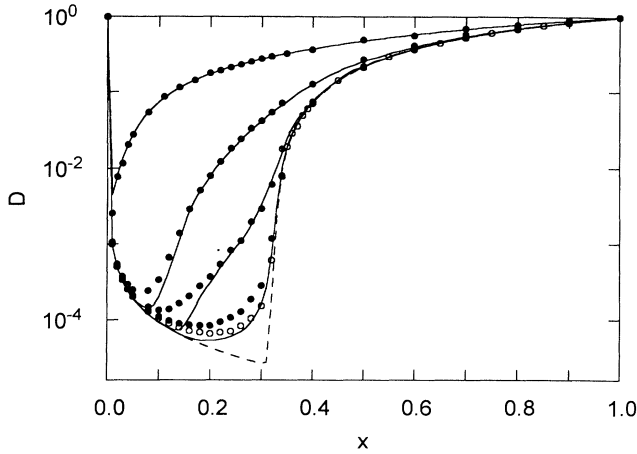


FIG. 1. Logarithmic plots of the normalized diffusion constant D as a function of the concentration x of lower energy sites, showing effects of long-range hopping on the trapping-to-percolation transition. Solid curves are the result of the long-range EPEMT, while solid circles are from the corresponding numerical simulations. Curves shown are for values of the inverse hopping length $\eta=2, 10, 18,$ and 26 . The nearest-neighbor EPEMT curve is shown as a dashed line, and the nearest-neighbor simulation data are shown as open circles.

intrinsic conductivity of the two pure systems is the same. The rate W_{12} out of the low-energy sites, however, is reduced due to detailed balance considerations. In all curves presented, we have taken $W_{12} = W_{21} \times 10^{-5}$. In all the solid curves, the length dependence of the rates takes the form $W_s^{\beta\alpha} = W(|s|; \varepsilon_\beta, \varepsilon_\alpha) = W_{\beta\alpha} \exp(-\eta|s|)$, where the parameter η determines the effective range of hopping. Solid curves are shown for values of $\eta=2, 10, 18,$ and 26 . The nearest-neighbor curve is shown as a dashed line. Results from a direct numerical simulation of the binary system for the same values of η are shown as solid circles. Numerical results for the nearest-neighbor case are shown as open circles.

As can be seen, the effective-medium curves reproduce the simulation results quite well except in the region where the conductivity minimum occurs. This minimum represents a transition between two essentially different conduction mechanisms. At small x , diffusion is “trap limited” with carriers spending a great deal of time in the isolated lower energy sites. At higher concentrations, it becomes easier to simply hop directly among the lower energy sites themselves, at which point conduction increases with the concentration of those sites. The transition is sharpest in the nearest-neighbor case, although the effective-medium theory clearly overestimates the sharpness of the transition. (The apparent cusp observed for $\eta=2$ is actually rounded in the simulations when a fine enough resolution in the concentration is used.)

One advantage of the effective-medium approach is that a given family of long-range percolation curves generated numerically (or analytically) for a set of length-dependent parameters can be used to efficiently calculate the effective-medium curves for a wide variety of energetic configurations of the constituent ions. As we have indicated in the formal development of the effective-

medium equations, the approach is not restricted to a binary system, so that more general site energy distribution functions can be used. Also, it is straightforward to use different length dependences for rates connecting sites of different types.

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APPENDIX: SOLUTION OF THE EFFECTIVE-MEDIUM EQUATIONS

In this Appendix we solve the energy-projected effective-medium equations and obtain an exact expression for the diffusion constant in terms of the effective-medium rates. To this end, we introduce a Fourier representation of the averaged probabilities with

$$p_k^\alpha(z) \equiv \sum_n e^{-ik \cdot n} p_n^\alpha(z), \quad (\text{A1})$$

defining the Fourier transform of $p_n^\alpha(z)$ at wave vector $\mathbf{k} = (k_1, \dots, k_d)$. The Fourier transform of Eq. (5) is

$$z p_k^\alpha(z) - P_k^\alpha(0) = \sum_\beta \Gamma_k^{\alpha\beta}(z) p_k^\beta(z), \quad (\text{A2})$$

in which $\Gamma_k^{\alpha\beta}(z) \equiv \sum_s e^{-ik \cdot s} \omega_s^{\alpha\beta}(z) - \delta_{\alpha\beta} \sum_{s,\gamma} \omega_s^{\gamma\beta}(z)$. The formal solution to (A2) can be written as follows:

$$p_k^\alpha(z) = \sum_\beta G_k^{\alpha\beta}(z) P_k^\beta(0), \quad (\text{A3})$$

in terms of the Green's function $G_k(z) = (z - \Gamma_k)^{-1}$, which is diagonal in \mathbf{k} but is an $N \times N$ matrix in the energy indices.

The diffusion constant D characterizes the asymptotic growth of the mean-square displacement of a transport particle as a function of time. This quantity is readily obtained from the solution given in (A3). A standard Tauberian theorem, along with (A1) implies that

$$\begin{aligned} 2dD &\equiv \lim_{t \rightarrow \infty} \frac{d}{dt} \langle n^2(t) \rangle = \lim_{z \rightarrow 0} \sum_{n,\alpha} z^2 n^2 p_n^\alpha(z), \\ &= - \lim_{z \rightarrow 0} \sum_{\alpha} z^2 \nabla_k^2 p_k^\alpha(z), \\ &= - \lim_{z \rightarrow 0} \sum_{\alpha,\beta} z^2 \nabla_k^2 [G_k^{\alpha\beta}(z) P_k^\beta(0)]. \end{aligned} \quad (\text{A4})$$

It turns out that we can restrict the derivatives in the last line of (A4) to G_k , because only the singular part of G_k at $k=0$ survives in the zero-frequency ($z=0$) limit. To show this we need to evaluate the derivatives of G_k and $P_k(0)$. Using the notation P for the vector with components $P_k^\beta(0)$, G for the operator with elements $G_k^{\alpha\beta}(z)$, and Γ for the operator with elements $\Gamma_k^{\alpha\beta}(z)$, the required derivatives can be written as follows:

$$z^2[\nabla_k^2 GP] = z^2[(\nabla_k^2 G)P + 2(\nabla_k G) \cdot (\nabla_k P) + G(\nabla_k^2 P)] . \quad (\text{A5})$$

Note also that by definition $GG^{-1} = 1$. Thus,

$$\begin{aligned} \nabla_k \{GG^{-1}\} &= 0 = \{\nabla_k G\}G^{-1} + G\{\nabla_k G^{-1}\} , \\ &= \{\nabla_k G\}G^{-1} - G\{\nabla_k \Gamma\} , \end{aligned} \quad (\text{A6})$$

which implies that

$$\nabla_k G = G\{\nabla_k \Gamma\}G . \quad (\text{A7})$$

It is then straightforward to extend this using the chain rule to obtain the result

$$\nabla_k^2 G = G\{\nabla_k^2 \Gamma + 2(\nabla_k \Gamma) \cdot G(\nabla_k \Gamma)\}G . \quad (\text{A8})$$

All terms in (A5) and (A8) involving the quantity $\nabla_k \Gamma$, including $\nabla_k G$ itself, vanish in the zero wave-vector limit due to the translational invariance of the effective medium. To see this, note that $\lim_{k \rightarrow 0} \nabla_k^{\alpha\beta} = -i \sum_s s \omega_s^{\alpha\beta}$. For centrosymmetric lattices, in which the negative of a lattice vector is also a lattice vector, the summand is odd insofar as the quantity $\omega_s^{\alpha\beta} = \omega_{-s}^{\alpha\beta}$ depends only on the magnitude of s . Thus, $\lim_{k \rightarrow 0} \nabla_k \Gamma_k = 0 = \lim_{k \rightarrow 0} \nabla_k G_k$. Hence we find that

$$\lim_{\substack{z \rightarrow 0 \\ k \rightarrow 0}} z^2 \nabla_k^2 (GP) = \lim_{\substack{z \rightarrow 0 \\ k \rightarrow 0}} z^2 [(\nabla_k^2 G)P + G(\nabla_k^2 P)] . \quad (\text{A9})$$

The last term on the right in (A9) will vanish in the zero-frequency limit, because

$$\lim_{z \rightarrow 0} z^2 G = \lim_{z \rightarrow 0} z^2 (z - \Gamma)^{-1} = 0 . \quad (\text{A10})$$

Combining these results we can reexpress (A4) in the form

$$\begin{aligned} 2dD &= - \lim_{\substack{z \rightarrow 0 \\ k \rightarrow 0}} \sum_{\alpha, \beta, \gamma, \delta} z^2 G_k^{\alpha\gamma} (\nabla_k^2 \Gamma_k^{\gamma\delta}) G_k^{\delta\beta} P_k^\beta(0) , \\ &= \lim_{z \rightarrow 0} \sum_{\alpha, \beta, \gamma, \delta} z g^{\alpha\gamma} \left[\sum_s s^2 \omega_s^{\gamma\delta} \right] z g^{\delta\beta} \sum_n P_n^\beta(0) , \end{aligned} \quad (\text{A11})$$

where we have introduced $g^{\alpha\beta} = \lim_{k \rightarrow 0} G_k^{\alpha\beta}$.

Let ρ_β denote the equilibrium population of states of energy ε_β corresponding to an arbitrary initial condition for one particle, i.e.,

$$\begin{aligned} \rho_\beta &\equiv \lim_{t \rightarrow \infty} \sum_n \langle P_n^\beta(t) \rangle = \lim_{\substack{t \rightarrow \infty \\ k \rightarrow 0}} \langle P_k^\beta(t) \rangle = \lim_{\substack{z \rightarrow 0 \\ k \rightarrow 0}} z P_k^\beta(z) , \\ &= \lim_{\substack{z \rightarrow 0 \\ k \rightarrow 0}} \sum_\alpha z G_k^{\beta\alpha}(z) P_k^\alpha(0) = \lim_{z \rightarrow 0} \sum_\alpha z g^{\beta\alpha} \sum_n P_n^\alpha(0) . \end{aligned} \quad (\text{A12})$$

For the particular localized initial condition $P_n^\alpha(0) = \delta_{n,m} \delta_{\alpha,\gamma}$, Eq. (A12) leads to the identity $\rho_\beta = \lim_{z \rightarrow 0} z g^{\beta\gamma}$ which, because of the assumed uniqueness of the equilibrium distribution, is independent of γ . Performing this limit on the two factors containing the elements of the matrix g appearing in Eq. (A11) we obtain

$$2dD = \sum_{\alpha, \beta, s} s^2 \omega_s^{\alpha\beta}(0) \rho_\beta , \quad (\text{A13})$$

which is the desired expression.

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