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Comments

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Effect of periodic boundary conditions in the calculation of ac conductivity for a one-dimensional percolation model

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The calculation of the ac hopping conductivity for a linear chain with random interruptions requires the solution of the hopping problem for an ordered segment of arbitrary *finite* length. This problem was treated recently by the author using periodic boundary conditions and, exactly, by Odagaki and Lax (OL) who considered properly terminated segments. It is shown that the results obtained using the OL definition of the averaged complex diffusion constant in the author's treatment for periodic boundary conditions coincide with the exact results to leading order for strong disorder, both at low and high frequencies. These new results are compared in detail with those of OL for the whole range of disorder. An important normalization correction is included in the author's earlier results, which were based on a different definition of the averaged diffusion constant. It is shown that the two definitions of the diffusion constant may lead to substantial quantitative differences.

Hopping on a linear chain lattice with random interruptions of the nearest-neighbor hopping rate along the chain has been studied extensively as a simple model for carrier or exciton transport and spectral diffusion in disordered systems. The carrier is assumed to hop between nearest neighbors only and the hopping rate has a constant value W_0 with probability p and vanishes with probability $1-p$ due, e.g., to random impurities or defects which interrupt the hopping path. It is clear that for $p\neq 1$ the infinite chain breaks up into a set of *ordered* chain segments, where a segment refers to a finite cluster of sites connected to each other by transfer rates W_0 which are regarded as bonds.

In particular, the frequency-dependent conductivity given by

$$\sigma(\omega)=\frac{ne^2}{k_B T}D(\omega), \tag{1}$$

where

$$D(\omega)=-\frac{\omega^2}{2}\left\langle \sum_{x,x'}(x-x')^2\tilde{\mathscr{P}}(x,i\omega\,|\,x')f(x')\right\rangle \tag{2}$$

is a generalized diffusion constant, has been studied recently by Heinrichs[1] and by Odagaki and Lax.[2] Here n is the density of effective carriers of charge e, x denotes a site position in the chain, and $f(x')$ is the equilibrium distribution function for the initial carrier position x'. $\tilde{\mathscr{P}}(x,u\,|\,x')$ is the Laplace transform of the probability $\mathscr{P}(x,t\,|\,x',0)$ of finding a carrier at a site x at time t if it was at x' at $t=0$ and is determined by the usual random-walk equation[3] for a fixed configuration of the random chain. The angular brackets in Eq. (2) denote configuration averaging. Two different forms for $f(x')$ have been suggested in the literature. Scher and Lax[3] assume $f(x')=\delta_{x',x_0}$, i.e., the carrier is initially placed in a localized state centered around a fixed site $x'=x_0$, while Odagaki and Lax[2] assume $f(x')=1/L$ (with L equal to the total number of sites in the system), i.e., the carrier is found initially with equal probability on any site (high-temperature limit). By performing the average over initial sites x' in the expression between angular brackets in Eq. (2) we obtain, for both forms of $f(x')$,

$$D(\omega)=-\frac{\omega^2}{2}\left\langle \sum_{x}(x-x_0)^2\tilde{\mathscr{P}}(x,i\omega\,|\,x_0)\right\rangle, \tag{2'}$$

where x_0 denotes the initial location of the carrier.

In the case where $f(x') = 1/L$, Eq. (2′) is obtained after interchanging averages over x' and over configurations in Eq. (2) and noting that the average in (2′) is independent of x_0 since the configuration averaging includes, in particular, averaging over the subensemble of realizations that differ only by the initial position of the carrier. Since $\mathscr{P}(x,t\,|\,x_0,0)$ and hence $\tilde{\mathscr{P}}(x,i\omega\,|\,x_0)$ are zero unless x and x_0 belong to the same segment, Eq. (2′) may be reduced to a weighted average over ordered segments (clusters) of different sizes:

$$D(\omega)=\left[\sum_{N=1}^{\infty} Y(N)\right]^{-1}\sum_{N=1}^{\infty} Y(N)D_N(\omega), \tag{3}$$

where N denotes the number of sites on a segment and the diffusion constant $D_N(\omega)$ for a segment is given by [with $D_1(\omega)\equiv 0$]

$$D_N(\omega)=-\frac{\omega^2}{2}\frac{1}{N}\sum_{x,x_0}^{N}(x-x_0)^2\tilde{P}(x,i\omega\,|\,x_0), \tag{4}$$

where $\tilde{P}(x,i\omega\,|\,x_0)$ is just $\tilde{\mathscr{P}}(x,i\omega\,|\,x_0)$ when x and x_0 belong to the same segment and the summation $(1/N)\sum_{x_0}^{N}\cdots$ results from averaging over the configurations corresponding to different initial positions along a segment of N sites. In Ref. 1 the diffusion constant $D_N(\omega)$ is calculated by applying periodic boundary conditions (PBC) for clusters with $N\geq 3$ (the pair cluster is treated exactly) and in Ref. 2 it is calculated exactly by considering properly terminated clusters. When PBC are used for an individual segment, all sites on it become equivalent and Eq. (4) reduces to a simple sum over sites x with the factor $1/N$ removed.[1]

Now, since an individual term in Eq. (2) may be regarded as depending either on an initial site for a carrier x' and on a distant hopping site x or, equivalently, on an initial site x' and on a hopping distance $x-x'$, it is as well suited for discussing a site percolation problem as for describing a bond percolation problem. This suggests the possibility of introducing two different configuration averages to define a generalized diffusion constant $D(\omega)$ of the form of Eq. (3). First, since one is actually dealing with a bond percolation problem (a bond corresponds to a transfer rate W_0 connecting two sites) a natural definition of $D(\omega)$ is obtained by averaging $D_N(\omega)$ with respect to the probability $R(N-1)$ for a *bond* to belong to a cluster of $N-1$ bonds (N sites) terminated by a broken bond at each end.[4] In this case

$$Y(N)\equiv R(N-1)$$
$$=(N-1)(1-p)^2p^{N-1}\,,\quad N\geq 2 \tag{5a}$$

$$Y(1)=1-p, \tag{5b}$$

where $Y(1)$ is the probability for a bond to be absent. One has the normalization condition[1]

$$1-p+\sum_{N=2}^{\infty}R(N-1)=1\;. \tag{6}$$

Note that since the $N=1$ term gives no contribution in the numerator of Eq. (3) [because $D_1(\omega)\equiv 0$], one might be led to conclude that the "no bond probability" $Y(1)$ should be omitted from the summation of weighting factors in the denominator of Eq. (3). However, this is incorrect because it would lead to an expression in which the effective weighting factor $Y(N)/\sum_{N=2}^{\infty}Y(N) = p^{-1}Y(N)$ for $D_N(\omega)$ would not vanish for $p = 0$ for $N = 2$, and hence the general property $\lim_{p\to 0}D(\omega)=0$ would not be obeyed. This shows that it is necessary to retain the $N = 1$ term in the denominator of Eq. (3) [where $\sum_{N=1}^{\infty}Y(N)=1$] in order to ensure that $D(\omega)$ vanishes when no bonds are present in the system.

Alternatively, one may define $D(\omega)$ as the average of $D_N(\omega)$ over the probability

$$Y(N)\equiv Q(N)=N(1-p)^2p^{N-1} \tag{7}$$

for any given *site* to belong to a cluster of N sites. $Q(N)$ satisfies the normalization condition

$$\sum_{N=1}^{\infty}Q(N)=1, \tag{8}$$

where $Q(1) = (1-p)^2$ gives the probability for a site to be disconnected from its nearest neighbors. As discussed above one must keep the contribution of isolated sites [$Q(1)$] in the summation of weighting factors in the denominator of Eq. (3) in order to preserve the proper behavior of $D(\omega)$ for $p\to 0$.

Heinrichs[1] used the definition of $D(\omega)$ in terms of the bond probability distribution $R(N-1)$, whereas Odagaki and Lax[2] used the definition in terms of the site distribution $Q(N)$. While both treatments yield the same frequency dependence for the real and imaginary parts of $D(\omega)$ at low and high frequencies, it would appear[2] that for strong disorder ($p\to 0$) the expansion coefficients for these two ranges remain finite in the work of Ref. 1 while going linearly to zero in that of Ref. 2. Unfortunately, the necessity of including the "no bond term" $Y(1)$ in the denominator of Eq. (3) was overlooked in Ref. 1 with the result that a factor p is missing [as shown by Eq. (6)] in the final expressions, Eqs. (31)–(33), (38), (42), (50), and (52), of Ref. 1. After correcting for this, the above expansion coefficients are also linear in p to leading order ($p < 1$), but their magnitude for $p \neq 0$ is only half that of the exact coefficients of Odagaki and Lax at the same order. However, we find that a direct comparison of the results of Refs. 1 and 2 for finite disorder ($p \neq 0$) is somewhat misleading because of the use of different definitions of $D(\omega)$ in the two treatments.

For the purpose of making a proper comparison with Ref. 2, we now discuss the results for PBC in the case where $D_N(\omega)$ is averaged with respect to the site distribution (7). Following the notation of Odagaki and Lax we write the low- and high-frequency expansions of $D(\omega)$ in the form [with $\tilde{D}(\omega) \equiv D(\omega)/W_0$]

$$\tilde{D}(\omega) = A\tilde{\omega}^2 + B\tilde{\omega}i + O(\tilde{\omega}^3), \quad \tilde{\omega} = \frac{\omega}{W_0} << 1 \tag{9}$$

$$\tilde{D}(\omega) = C + \frac{B'}{\tilde{\omega}}i + O(\tilde{\omega}^{-2}), \quad \tilde{\omega} >> 1. \tag{10}$$

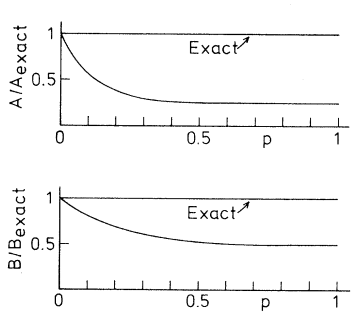

FIG. 1. Comparison of coefficients of the low-frequency expansion of the generalized diffusion constant with the exact results of Odagaki and Lax (Ref. 2) as a function of disorder. A and B are defined in the text.

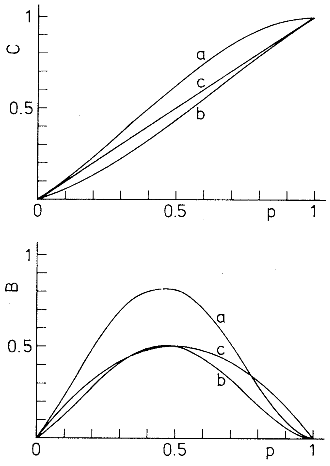

FIG. 2. Comparison of coefficients of the high-frequency expansion of the generalized diffusion constant with exact results of Odagaki and Lax (Ref. 2) as a function of disorder. Curves a: coefficients C and B' defined in the text. Curves b: coefficients $C \equiv C_R$ and $B' \equiv B_R'$ obtained in the case of the bond-averaged diffusion constant (Ref. 1). Curves c: exact results of Odagaki and Lax (Ref. 2).

Using the detailed results of Ref. 1 for $D_N(\omega)$ and performing the average in Eq. (3) with respect to $Y(N) = Q(N)$ we obtain the final expressions

$$A = \tfrac{1}{4}p(1-p)^2 \left[1 - \frac{1}{8p}\left(-\tfrac{1}{15}S_5 - \tfrac{1}{18}S_3 + \tfrac{1}{4}S_2 + \tfrac{31}{90}S_1 \right) \right], \tag{11}$$

$$B = \tfrac{1}{2}p(1-p)^2 \left[1 + \frac{1}{12p}(S_3 - S_1) \right], \tag{12}$$

$$C = p(1+p-p^2), \tag{13}$$

$$B' = p(2+9p)(1-p)^2, \tag{14}$$

where

$$S_n = \sum_{N=3}^{\infty} N^n p^{N-1}, \tag{15}$$

whose values for various n have been tabulated in Ref. 1. These expressions should be compared with the exact results of Odagaki and Lax, namely[2] $A_{\text{exact}} = 4^{-1}(1-p)^{-4}p(1+p)^2$, $B_{\text{exact}} = 2^{-1}(1-p)^{-2}p$, $C_{\text{exact}} = p$, and $B'_{\text{exact}} = 2p(1-p)$. The most important conclusions follow at once from Eqs. (11)–(14): (i) to leading order in p for $p < 1$ we get the values $A = p/4$, $B = p/2$, $C = p$, and $B' = 2p$ which coincide with the exact results at the same order in p. We note that to leading order the corresponding corrected coefficients

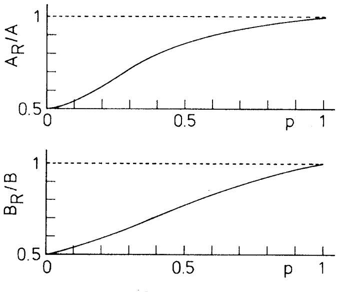

FIG. 3. Comparison of the low-frequency expansion coefficients for the bond- and site-averaged diffusion constants. A and B are the coefficients defined in the text and A_R and B_R are those obtained in Ref. 1.

found in Ref. 1 are [using a subscript R to indicate that they relate to an average with respect to the bond distribution (5a),(5b)] $A_R = p/8$, $B_R = p/4$, $C_R = p/2$, and $B'_R = p$. Their magnitude is seen to be half that of the coefficients A, B, C, and B' evaluated at the same order, which indicates already a significant difference between the two averaging procedures. (ii) for p close to unity the coefficients A and B show the same critical behavior as A_{exact} and B_{exact}, namely $A \sim (p-p_c)^{-4}$, $B \sim (p-p_c)^{-2}$, $p_c = 1$, and the coefficients C and B' coincide with the corresponding exact values at $p=1$.

In Figs. 1 and 2 we compare the above expressions, Eqs. (11)–(14), with the exact results of Odagaki and Lax for the whole range of values of the order parameter $0 \leq p \leq 1$. Also, in order to illustrate quantitatively the differences between the two averaging procedures discussed above, we compare in Figs. 2 and 3 the expressions (11)–(14) with the corrected results of Ref. 1.[5]

In conclusion, we have presented a proper comparison of Heinrichs's calculation of ac conductivity for the interrupted chain model and the exact calculation of Odagaki and Lax, in the low- and high-frequency regimes. Such a comparison is of interest since the main purpose of the exact solution of Odagaki and Lax is to provide a standard against which approximate treatments of percolation conduction problems can be tested in order to assess their range of validity as well as their relative merits.[2] At low frequencies (long times) the use of PBC is least reliable for $p \to 1$ because, on the one hand, cluster boundary effects are expected to play an important role in this case and, on the other hand, the dominant contribution to $D(\omega)$ comes from clusters of larger and larger size whose boundaries are not properly described by imposing PBC. At high frequencies (short times), end effects are expected to be relatively unimportant for any cluster size. This explains why in this case the deviations of the above results from the exact ones of Ref. 2 are generally small for any disorder. Finally, we have compared the results based on PBC for two different definitions of the averaged diffusion constant and obtained important differences, particularly for strong disorder.

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[1] J. Heinrichs, Bull. Am. Phys. Soc. 25, 369 (1980); Phys. Rev. B 22, 3093 (1980).

[2] T. Odagaki and M. Lax, Phys. Rev. Lett. 45, 847 (1980).

[3] See, e.g., Appendix B of H. Scher and M. Lax, Phys. Rev. B 7, 4491 (1973).

[4] In this connection the last sentence before Eq. (28) of Ref. 1 should be clarified to read "Since the probability of finding a segment of connected bonds of length N ($N+1$ sites) is $(1-p)^2p^N$, the probability $R(N)$ for any given bond to belong to a segment of length N is . . . ."

[5] The factor $p^2(3-2p)$ in the first term of the square bracket in Eq. (38) and in the second term of the square bracket in Eq. (42) of Ref. 1 is incorrect and should be replaced by $p(2-p)$.