Hopping Conduction on Aperiodic Chains

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The dynamic conductivity of one-dimensional hopping systems with aperiodically distributed transition rates is studied. The low-frequency behavior is shown to be regular or singular depending on the specific substitution which generates the aperiodicity. Explicit formulas are given for three cases with different spectral measures of the transition-rate sequences. We give general high-frequency expansions that are valid whenever the correlation functions exist. A numerical calculation of the conductivity in the range of intermediate frequencies by a decimation procedure is also included.

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Deterministic aperiodic structures, representing an intermediate situation between the random and periodic ones, are a topic much studied at present, especially since the quasicrystalline order was discovered experimentally.[1] The theoretical investigation of one-dimensional quasicrystals produced nontrivial results concerning the electronic and phononic spectrum,[2] magnetic properties, and phase transitions in the Ising model.[3] At the same time, different systems consisting of aperiodic sequences of some physical parameters (atomic masses, coupling constants, etc.), generated by finite automata, has been approached.[4]

This Letter treats the hopping-transport properties of aperiodic chains. Recent studies concerning hopping on one-dimensional systems pointed out the importance of the distribution of their constituents in establishing the qualitative behavior of the response to external fields. So the expression for the low-frequency ac conductivity, which is a problem of first importance in hopping, is regular for periodic chains and becomes nonanalytic when the transition rates are distributed randomly.[5] Our aim is to complete this picture, analyzing the intermediate case represented by the deterministic aperiodic systems, and at the same time to identify the basis of the different frequency dependences.

The model considered here consists of a binary aperiodic sequence of transition rates between isoenergetic sites. The sequence is generated by the use of a substitution procedure. The structure is completely determined by our giving a finite alphabet, the initial element, and a specific rule for the replacement of each letter of the alphabet with a finite word. If the substitution is applied iteratively, a semi-infinite aperiodic chain is built up.

The hopping conductivity will be calculated with use of the rate equations for the average number of carriers ρ_n localized at the site x_n. One assumes that the transition rates satisfy detailed balance in the presence of the external electric field. The linearization of the rate equations gives rise to the so-called "resistance network analogy" introduced by Miller and Abrahams.[6] In this approach the hopping current is given by the time derivative of the dipole moment:

$$j=\lim_{L\to\infty}\frac{e}{L}\sum_n \dot{\rho}_n x_n, \tag{1}$$

where e is the electric charge and L the length of the chain. For one-dimensional systems the formalism is detailed elsewhere.[7]

Let d_n and w_n denote the distance and the transition rate between the sites n and $n+1$, and ω the frequency of the external electric field. Then, if we linearize with respect to the field E, for the model under consideration the conductivity can be written as

$$\sigma(\omega)=\frac{1}{E}\lim_{L\to\infty}\frac{1}{L}\sum_n I_n d_n, \tag{2}$$

where the "elementary currents" I_n, representing the charge flow between the sites n and $n+1$, are the solution of the equations[7]

$$(i\omega/w_n+2)I_n=I_{n+1}+I_{n-1}+i\omega e E d_n. \tag{3}$$

First we shall study the influence of the aperiodicity on the low-frequency conductivity. In order to simplify our discussion we shall consider at this stage only equidistant sites ($d_n=1$). The infinite chain is considered as the limit of a finite chain of N sites with periodic boundary conditions. Then, by our making use of the lattice Fourier transformation of Eq. (3), the dynamic conductivity can be expressed as a formal fluctuation expansion[5]:

$$\sigma(\omega)=\sigma(0)+\frac{i\omega}{m_{-1}^2}\lim_{N\to\infty}\sum_{q\neq 0}\frac{|S_N(q)|^2}{2(1-\cos q)+i\omega m_{-1}}+\frac{\omega^2}{m_{-1}^2}R(\omega), \tag{4}$$

where

$$m_{-1}=\lim_{N\to\infty}\frac{1}{N}\sum_n w_n^{-1},\quad \sigma(0)=1/m_{-1},$$

and $S_N(q)$ is the Fourier transform of the fluctuations $\delta_n = w_n^{-1} - m_{-1}$:

$$S_N(q) = N^{-1} \sum_n e^{iqx_n} \delta_n \quad (q = 2\pi k/N,\ k = 0, \ldots, N-1).$$

The remainder $R(\omega)$ represents the sum of all terms containing fluctuation correlations of order higher than 2. When the limit $N \to \infty$ is performed, the sum $\sum_q |S_N(q)| \cdots$ goes into the integral $\int dM(q) \cdots$, where $dM(q)$ is called the spectral measure of the fluctuation sequence.[4] If the first two terms in Eq. (4) are sufficient to describe the qualitative frequency dependence of the conductivity in the limit $\omega \to 0$, then it comes out that the asymptotic behavior is controlled by the spectral measure $dM(q)$.

The low-frequency asymptotics of the random and periodic binary chains are already known[5]: $\mathrm{Re}\sigma(\omega) - \sigma(0)$ behaves like $\omega^{1/2}$ in the first case and like ω^2 in the second one, which reveals strong differences as regards the analyticity at $\omega = 0$. This can be explained in terms of spectral measures: In the random case $dM(q) = dq$ (Lebesgue measure), while the spectral measure of the periodic chain is a sum of equidistant, equally weighted δ functions.

The question of interest is how the aperiodic chains generated by various substitutions will behave. We choose three examples for which the corresponding substitutions yield different spectral measures. For the beginning, let us focus our attention on the more familiar Fibonacci sequence. In this case, we have a two-letter alphabet $\{A,B\}$ and the substitution rule $A \to AB$, $B \to A$ so that, starting with A, the resulting chain is $ABAAB\ldots$ We notice that [since $q = 0$ is excluded in the sum of Eq. (4)] for the calculation of $S_N(q)$ in the case of binary chains we need to know only the sum over one species of transition rates (let us say A), i.e.,

$$S_N^A(q) = (\delta_A/N) \sum_n e^{iqx_n^A},$$

where x_n^A denote the left ends of the A-type bonds. For the Fibonacci sequence $x_n^A = n + [(n+1)/\tau]$, where $[x]$ represents the integer part of x and τ is the golden mean. The calculation of $S_N(q)$ can be carried out in a similar way to the structure-factor calculation for the Fibonacci quasicrystal (see, for instance, Duneau and Katz,[8] Elser,[8] and Kalugin, Kitaev, and Levitor[8]). The resulting spectral measure is a sum of weighted δ functions centered at the points $q_{nm} = 2\pi(n\tau + m)/\tau^2$ (n and m are any integers), the weights being $2(1 - \cos q_{nm})/(q_{nm} - 2\pi m)^2$. Then the integral arising from the sum in (4) can be performed and the following low-frequency conductivity formula is obtained:

$$\sigma(\omega) = \sigma(0) + \frac{2i\omega}{m_{-1}^2} \left(\frac{\Delta\tau}{\pi} \right)^2 \sum_{n=1}^{\infty} \frac{1}{n^2} \frac{\sin^2(\pi n/\tau)}{4\sin^2(\pi n/\tau) + i\omega m_{-1}} + \frac{\omega^2}{m_{-1}^2} R(\omega), \tag{5}$$

where $\Delta = |\delta_A - \delta_B|$. The above formula shows that the dominant small-ω dependence of $\mathrm{Im}\sigma$ is analytic and similar to the periodic case (i.e., proportional to ω). On the other hand, numerical calculations reveal that, for $\mathrm{Re}\sigma$, the low-frequency dependence is of the form $\omega^2(1 - \mathrm{const}\ln\omega)$. The nonanalytic dominant behavior at $\omega = 0$ is made evident by the fact that the expression

$$\omega^{-2}[\mathrm{Re}\sigma(\omega) - \sigma(0)]\,|_{\omega=0} \simeq \sum_{n=1}^{\infty} \frac{1}{n^2} \frac{1}{\sin^2(n\pi/\tau)} \tag{6}$$

diverges. (In order to prove this, it should be observed that whenever n equals a Fibonacci number, the corresponding term in the sum is greater than $1/\pi^2$.) The calculation of the higher-order terms in the expansion (5) can be carried out in the same way. In the third order, in the limit $\omega \to 0$, the imaginary part is dominated by, while the real part is similar to, the second-order term. Hence, the third-order contribution in (5) does not change the dominant asymptotic behavior given by the previous one.

For the next part of our discussion we choose a well-known case with purely singular continuous spectral measure, namely the Thue-Morse chain. This chain is generated by the same alphabet and initial element as in the previous case, but the substitution rule is $A \to AB$, $B \to BA$. We note that after p consecutive substitutions the chain is a word of $N = 2^p$ letters. If we take into account that the fluctuations satisfy the relation $\delta_n = \delta_{2n} = -\delta_{2n+1}$, the explicit form of $|S_N(q)|$, which can be obtained by recurrence, is the following:

$$|S_N(q)| = \tfrac{1}{2}\Delta \left| \prod_{n=1}^{p} \sin q 2^{p-n-1} \right|. \tag{7}$$

Denoting by g_N the sum over q in (4) for the finite chain of length N, the following recursion relation can be proved by the use of (7) and some elementary algebra:

$$g_{2N}(s) = \tfrac{1}{8}\Delta^2 - 4s(1+2s) g_N(4s^2 + 4s), \tag{8}$$

$$s = i\omega m_{-1}/4.$$

An expansion in powers of s, performed at the fixed point (infinite N), gives

$$g_\infty(s) = \tfrac{1}{4}\Delta^2\{\tfrac{1}{2} - 2s + 28s^2 + O(s^3)\}, \tag{9}$$

and introducing (9) in (4) one obtains

$$\sigma(\omega) = \sigma(0) + i\omega \frac{\Delta^2}{8m_{-1}^2} + \omega^2 \frac{\Delta^2}{8m_{-1}} + O(\omega^2). \tag{10}$$

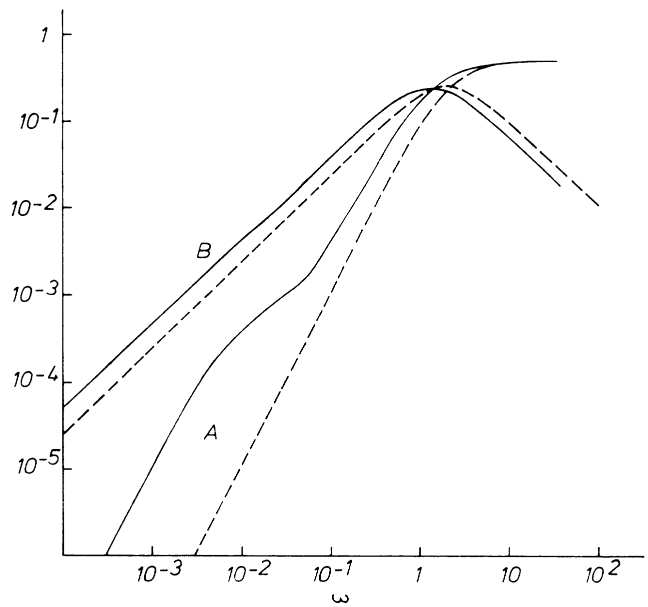

FIG. 1. $\mathrm{Re}\sigma(\omega)-\sigma(0)$ (curves A) and $\mathrm{Im}\sigma(\omega)$ (curves B) as functions of frequency for the Thue-Morse and periodic chain (solid and dashed lines, respectively). The transition rates are $w_A=1$, $w_B=0.01$, and ω is in units of w_A.

The only extra contribution to the ω^2 coefficient comes from the term of $R(\omega)$ containing the third-order fluctuation correlation, but again the remainder does not change the dominant small-ω dependence.

The above expression shows that the qualitative low-frequency behavior of the Thue-Morse chain is similar to that of the periodic chain. Nevertheless, at larger frequencies this similarity is lost. This can be seen in Fig. 1 which represents numerical results for $\sigma(\omega)$. They are obtained by our solving Eqs. (3) by the decimation procedure. For comparison, the dynamic conductivity of the periodic chain $w_A, w_B, w_A, w_B, \ldots$ is also shown. One sees that, at low and high frequencies, $\sigma(\omega)$ exhibits for both chains a similar ω behavior, but strong differences are manifested in the intermediate range.

Returning to the problem of low-frequency dependence, we find that interesting properties are exhibited by the Rudin-Shapiro chain. This is built up by a four-letter alphabet $\{A,B,C,D\}$, the substitution rule $A\to AB$, $B\to AC$, $C\to DB$, $D\to DC$, and in the resulting sequence replacement of each A or B with A and C or D with B. In this way a binary chain is obtained. From this construction it follows that

$$\delta_n=\delta_{4n}=\delta_{4n+1}=(-1)^n\delta_{4n+2}=-(-1)^n\delta_{4n+3}.$$

This relation gives rise to a recurrence formula for the second-order fluctuation correlations, which indicates that in the limit $N\to\infty$

$$N^{-1}\sum_n \delta_n\delta_{n+p}\sim\delta_{p,0}N^{-1}\sum_n\delta_n^2. \tag{11}$$

By the use of this result in the expression for $|S_N(q)|$, one concludes that, in this case, the spectral measure is $dM(q)=dq$ so that the integral over q can be performed giving an $\omega^{1/2}$ dependence, as for the disordered chain. Since the third-order fluctuation correlation vanishes, the third term in (4) does not contribute to the series. The higher terms give powers of ω greater than $\frac{1}{2}$ and, therefore, the low-frequency dependence of the conductivity is randomlike, i.e., nonanalytic. Generalizing our analysis, we can say that whenever the second-order correlation function is diagonal, the corresponding chain behaves like a random one.

The rest of this Letter is devoted to the high-frequency dependence of $\sigma(\omega)$, which is a much simpler problem because Eqs. (3) permit an immediate construction of an asymptotic series in powers of $1/\omega$. With the iterative procedure presented in Ref. 7, adapted to the chains with arbitrary-length bonds, the real and imaginary parts of the $1/\omega$ expansion read

$$\begin{aligned}\mathrm{Re}\sigma(\omega)&=m_1-b/\omega^2+O(1/\omega^4),\\ \mathrm{Im}\sigma(\omega)&=a/\omega+O(1/\omega^3),\end{aligned}\tag{12}$$

where

$$\begin{aligned}m_1&=\lim_{L\to\infty}\frac{1}{L}\sum_n d_n\bar{w}_n,\quad \bar{w}_n=d_nw_n;\quad a=\lim_{L\to\infty}\frac{1}{L}\sum_n(\bar{w}_n^2-\bar{w}_n\bar{w}_{n+1});\\ b&=\lim_{L\to\infty}\frac{1}{L}\sum_n\bar{w}_n\{4[\bar{w}_nw_n-\bar{w}_{n+1}(w_n+w_{n+1})]+\bar{w}_n(w_{n-1}+w_{n+1})+2w_{n+1}w_{n+2}\}.\end{aligned}\tag{13}$$

The values of the coefficients m_1, a, and b depend on the specific chain, but the high-frequency behavior of the ac conductivity expressed by Eq. (12) is universal, in the sense that it is valid for any type of chain: random, deterministic aperiodic, or periodic. Obviously, the asymptotic series makes sense only for those chains for which the coefficients exist, i.e., the appearance frequencies of the works occurring along the chain converge as L goes to infinity. Once the generating procedure is known, these frequencies can be calculated.[9] For instance, in the Fibonacci case the resulting coefficients are

$$m_1=\frac{\tau d_A\bar{w}_A+d_B\bar{w}_B}{\tau d_A+d_B},\quad a=\frac{2(\bar{w}_A-\bar{w}_B)^2}{\tau d_A+d_B},\quad b=a[2w_B+(2-1/\tau)w_A]. \tag{14}$$

In the above relations, the distances d_A and d_B are not specified. The Fibonacci quasicrystal corresponds to $d_A = \tau d_B$.

In conclusion, the influence of the aperiodicity of the transition-rate distribution on the dynamic hopping conduction of one-dimensional systems has been investigated. We find that, for our model, the low-frequency conductivity depends on the spectral measure of the sequence of transition rates on the chain. A sufficient condition which determines a specific behavior is the following: Any deterministic sequence whose second-order fluctuation correlation is diagonal [see Eq. (11)] will have a Lebesgue spectral measure and, consequently, a randomlike behavior. This is the case of the Rudin-Shapiro chain. The Thue-Morse sequence, described by a purely singular continuous spectral measure, behaves crystallike in the low-frequency limit. An intermediate situation is revealed by the Fibonacci sequence: In the limit $\omega \rightarrow 0$, the imaginary part of the conductivity is crystallike, while the real part is nonanalytic ($\sim \omega^2 \ln \omega$), but different from the typical randomlike behavior ($\sim \omega^{1/2}$). This frequency dependence is obtained here for a discrete spectral measure but one expects that nontrivial behaviors might result from the continuous part of the spectral measures. Finally, the high-frequency dependence of $\sigma(\omega)$ is shown to be independent of the transition-rate distribution.

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[1]D. Shechtman, I. Blech, D. Gratias, and J. W. Cahn, Phys. Rev. Lett. **53**, 1951 (1984).

[2]M. Kohmoto and J. R. Banavar, Phys. Rev. B **34**, 563 (1986); J. M. Luck and D. Petritis, J. Stat. Phys. **42**, 289 (1986).

[3]Y. Achiam, T. C. Lubensky, and E. W. Marshall, Phys. Rev. B **33**, 6460 (1986).

[4]J. P. Allouche and M. Mendes-France, J. Phys. (Paris), Colloq. **47**, C3-63 (1986); F. Axel, J. P. Allouche, M. Kleman, M. Mendes-France, and J. Peyrière, J. Phys. (Paris), Colloq. **47**, C3-181 (1986).

[5]R. Zwanzig, J. Stat. Phys. **28**, 127 (1982); S. Alexander, J. Bernasconi, W. R. Schneider, and R. Orbach, Rev. Mod. Phys. **53**, 175 (1981).

[6]A. Miller and E. Abrahams, Phys. Rev. **120**, 745 (1960).

[7]A. Aldea and M. Dulea, J. Phys. C **19**, 4055 (1986).

[8]M. Duneau and A. Katz, Phys. Rev. Lett. **54**, 2688 (1985); V. Elser, Phys. Rev. B **32**, 4892 (1985); P. A. Kalugin, A. Yu. Kitaev, and L. Levitov, Pis'ma Zh. Eksp. Teor. Fiz. **41**, 119 (1985) [JETP Lett. **41**, 145 (1985)].

[9]J. Peyrière, J. Phys. (Paris), Colloq. **47**, C3-41 (1986).