Correlation effects in hopping conductivity

Kuljinder S. Chase and D. J. Thouless
Department of Physics (FM-15), The University of Washington, Seattle, Washington 98195
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We introduce an alternative mapping of the hopping problem onto a random resistor network, which takes into account correlations among the occupation probabilities of different sites. The effect of correlations on the low-temperature conductivity of one-dimensional chains in the nearest-neighbor and variable-range-hopping cases is investigated by approximation of the network conductances by the critical conductance at the percolation threshold. Correlation effects are found to lead to increased activation energies and a mesoscopic behavior which is much closer to experimental data than that predicted by mean-field theory.

I. INTRODUCTION

Thermally assisted hopping of localized charge carriers is thought to be the dominant transport mechanism in disordered systems, when the Fermi level lies far below the mobility edge. The matrix element for hopping between a given pair of levels is essentially given by the product of a tunneling factor $\exp(-\alpha|\mathbf{R}_{ij}|)$ and a Boltzmann factor $\exp[-(E_i-E_j)/kT]$. At high temperatures the tunneling factor dominates and nearest-neighbor hopping pertains, while at low-temperatures carriers prefer to jump longer distances in order to find states closer in energy, resulting in a temperature-dependent activation energy, as was first proposed by Mott.[1] The low-temperature dc conductivity has the form

$$\sigma(T)\sim\exp[-(T_0/T)^{1/4}] \tag{1.1}$$

and has been experimentally observed in a wide variety of materials over fairly large temperature intervals.

Theoretical treatments of the low-temperature hopping problem revolve around the equivalent-resistor-network mapping of Miller and Abrahams,[2] and its subsequent percolative analysis by Ambegaokar, Halperin, and Langer[3] (to be referred to as AHL). Because of the competition between the activation-energy term and the tunneling factor the states involved are mostly of the order of 5 kT–10 kT distant from the Fermi energy, both below and above it; it is generally assumed that the average density of states is constant within this energy range that spans the Fermi energy.

An ingredient of this mapping that has been challenged[4] is the assumption that the occupation probabilities of different sites are uncorrelated. Intuitively correlations are expected to be relevant, because a carrier may only hop to another site if that site is unoccupied. This is particularly important for hops across the Fermi level and back, because once a particle has been excited to a state above the Fermi level it may only come back down to a different site if another carrier is excited. Richards[5] has demonstrated the importance of this effect, for the case of an ordered binary chain at half-filling with nearest-neighbor hopping, and shown that the activation energy is raised above the mean-field expectation. We shall discuss this model in more detail later as it clearly demonstrates the role of correlations.

In this paper we examine the effects of correlations on the conductance by mapping the problem onto a resistor network in which the nodes are many-particle configurations rather than single-particle states so that the occupation numbers are strictly one or zero. This mapping has the additional virtue that interactions between carriers can easily be incorporated, though we shall not consider them further in this paper.

Because we consider many-particle configurations rather than single-particle states the networks are complex and our study is restricted to rather short one-dimensional chains, where we find that correlations produce substantial changes in both nearest-neighbor and variable-range hopping, especially at the mesoscopic level where the structure in the conductance of a single sample as a function of the chemical potential[6] is found to arise for different reasons than in mean-field theory.[7] In addition no flat regions are observed and the results are much closer to experimental data than those predicted by mean-field theory.

II. A MANY-PARTICLE RANDOM RESISTOR NETWORK

The mapping of the hopping problem onto a random resistor network introduced by Miller and Abrahams[2,3] neglects correlations among the occupation probabilities of different sites. Site-site correlation functions are replaced by products of Fermi functions, which is only correct in the absence of an external field, when the system is in equilibrium. In the presence of an external electric field a current may flow between the two sites, and as discussed in the Introduction the occupation probabilities of the sites are expected to be correlated.

It turns out that by considering many-particle configurations instead of single-particle states it is possible, using arguments analogous to those of AHL, to map the problem onto an alternative resistor network in which correlations are naturally accounted for and in which in-

teractions between carriers can also be easily incorporated.

Following AHL we consider a system consisting of strongly localized single-particle states with energies specified by some given distribution. Each state is restricted to having an occupancy of 0 or 1. We assume that the carriers are noninteracting but the arguments given below are unchanged by interactions as long as one replaces changes in site energies by changes in total energy. We take the intrinsic transition rate between a given pair of sites to be given by

$$\gamma_{ij}=\begin{cases}\gamma_0 e^{-\alpha|\mathbf{R}_{ij}|-(E_i-E_j)/kT} & \text{for } E_j>E_i\\ \gamma_0 e^{-\alpha|\mathbf{R}_{ij}|} & \text{for } E_j<E_i,\end{cases} \tag{2.1}$$

where $\alpha/2$ is the inverse localization length and the E_i are single-particle energy levels. This satisfies the principle of detailed balance, which implies that in equilibrium

$$\gamma_{ij}=\gamma_{ji}e^{(E_i-E_j)/kT}. \tag{2.2}$$

In the presence of an external field ε detailed balance no longer holds and the transition rates are modified to

$$\hat{\gamma}_{ij}=\gamma_{ij}e^{-e\boldsymbol{\varepsilon}\cdot\mathbf{R}_{ij}/kT}, \tag{2.3}$$

which for small fields can be linearized to give

$$\hat{\gamma}_{ij}=\gamma_{ij}+\delta\gamma_{ij}=\gamma_{ij}-\gamma_{ij}e\boldsymbol{\varepsilon}\cdot\mathbf{R}_{ij}/kT. \tag{2.4}$$

We shall work in the small field limit and keep only terms up to $O(\varepsilon)$.

Consider two configurations of the system I and J which are connected by the hop of a single particle between some pair of sites i and j. Configurations are specified by a set of occupation numbers $\{n_1^I, n_2^I, \ldots, n_N^I\}$, each of which can be zero or one. The net rate at which charge flows between these two configurations is given by

$$\mathcal{J}_{IJ}=\hat{P}_I\hat{\gamma}_{ij}-\hat{P}_J\hat{\gamma}_{ji}, \tag{2.5}$$

where $\hat{P}_I$ is the probability of finding the system in configuration I in the presence of a small field ε. We shall write this as

$$\hat{P}_I=P_I+\delta_I, \tag{2.6}$$

where δ_I is the change in the configurational probability and P_I is the zero-field or equilibrium-configuration probability which is simply given by

$$P_I=\frac{e^{-(E_I-\mu N_I)/kT}}{Z}. \tag{2.7}$$

Here E_I is the total energy of configuration I, N_I is the number of carriers in state I, μ is the chemical potential, and Z the grand canonical partition function. We shall not need to evaluate δ_I explicitly in what follows. Substituting for $\hat{P}$ and $\hat{\gamma}$ in Eq. (2.5) leads to

$$\mathcal{J}_{IJ}=\frac{P_I\gamma_{ij}}{kT}\left[\boldsymbol{\varepsilon}\cdot\mathbf{R}_{ij}+kT\left[\frac{\delta_I}{P_I}-\frac{\delta_J}{P_J}\right]\right], \tag{2.8}$$

where we have used the fact that $P_I\gamma_{ij}$ equals $P_J\gamma_{ji}$. The quantity $kT\delta_I/P_I$ is just the change $\delta\mu_I$ in the local chemical potential of configuration I, so

$$\mathcal{J}_{IJ}=\frac{P_I\gamma_{ij}}{kT}(\boldsymbol{\varepsilon}\cdot\mathbf{R}_{ij}+\delta\mu_I-\delta\mu_J). \tag{2.9}$$

The term in parentheses is the total potential difference between configurations I and J, so we can consider them to be linked by a conductance

$$G_{IJ}=\frac{P_I\gamma_{ij}}{kT}. \tag{2.10}$$

Thus we can replace our system by a resistor network in which the nodes represent many-particle configurations and the resistors hops between these. This should be contrasted with the Miller and Abrahams mapping where the conductances connect single-particle states. The equivalent network just described is necessarily more complex than the mean-field one, as the number of configurations increases exponentially with the number of single-particle states.

III. THE ORDERED BINARY CHAIN

To illustrate the many-particle network discussed above consider an ordered binary chain consisting of alternating particle and hole states, with nearest-neighbor hopping only. The effect of correlations can be demonstrated by taking a pair of particle-hole states connected in a ring geometry. Consider first the case where there are two particles in the system. There are six possible configurations (1100), (1010), (1001), (0110), (0101), and (0011), whose occupation probabilities P_1 to P_6 are given by

$$\begin{aligned}&P_1=P_3=P_4=P_6=\frac{e^{-E/T}}{Z},\\ &P_2=P_1e^{-E/T},\\ &P_5=P_1e^{E/T};\end{aligned} \tag{3.1}$$

E is the energy difference between particle and hole states. For this simple example one can explicitly evaluate the changes in these probabilities when a small field ε is applied by solving the current conservation equations. One obtains

$$\begin{aligned}&\delta_1=\delta_6=-\delta_3=-\delta_4,\\ &\delta_2=\delta_5=0,\\ &\delta_1=\frac{(r-1)}{(r+1)}P_1\varepsilon,\end{aligned} \tag{3.2}$$

where $r=\exp(-E/T)$. The site-site correlation function

$$\langle n_1n_2\rangle-\langle n_1\rangle\langle n_2\rangle=-P_1+\delta_1 \tag{3.3}$$

picks up a contribution proportional to the applied field through δ_1. The constant part arises because we have kept the number of particles fixed, and vanishes in the grand canonical ensemble where the $\langle n_j\rangle$ are given by the Fermi distribution. Thus the external field induces

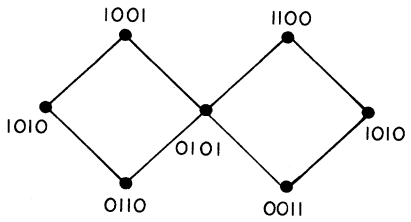

FIG. 1. The equivalent resistor network for a four-site system with two particles and periodic boundary conditions.

correlations between sites, which may not be neglected since we have kept terms of order ε throughout.

The equivalent network for this system is shown in Fig. 1 and has a conductance

$$G \sim \exp(-2E/kT) . \tag{3.4}$$

This result should be compared with the mean-field prediction which gives an activation energy of only E. The conductance is dominated by the two-particle–two-hole Boltzmann factor because both particles must be excited in order to transport a carrier around the ring. Despite their lower probability in equilibrium, one- or three-particle configurations have a lower energy cost, requiring an activation energy of only E, since only a single particle need be excited at any given time, and the cost of having these is simply $\exp(-\mu/kT)$ and $\exp[-(E-\mu)/kT]$, respectively. The total conductance of the ring is given by the sum of these three terms, and can be written as

$$G \sim \exp[-(\tfrac{3}{2}E - |\tfrac{1}{2}E - \mu|)/kT] . \tag{3.5}$$

An interesting feature of this result is that the conductance is not independent of the chemical potential, as mean-field theory predicts, when the Fermi level lies between 0 and E. A comparison of the predictions of the two theories calculated by solving the relevant network equations is shown in Fig. 2. Note that the peak value of the conductance when correlations are taken into account is lower than that predicted by mean-field theory, and the peaks are rounder than the valley. We shall come back to these points when we discuss random chains.

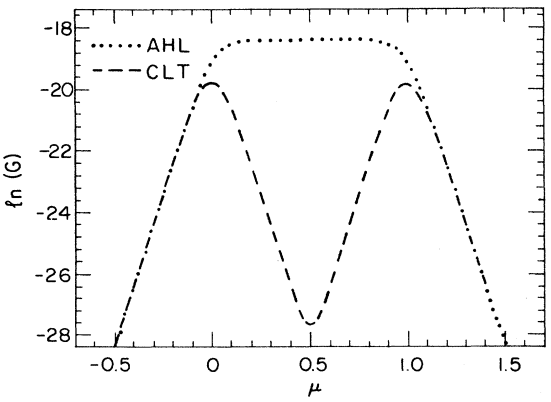

FIG. 2. The conductance of an ordered binary chain as a function of the chemical potential as predicted by mean-field theory (AHL) and the effect of including correlations (CLT). Units are arbitrary.

IV. RANDOM SYSTEMS

One does not expect the temperature dependence of the conductance in a large random system to be qualitatively altered by the inclusion of correlation effects. Mean-field theory is expected to be correct for pure particle or pure hole transport, and this provides a lower bound on the conductance. Correlations are expected to suppress the conductance so mean-field theory for both particle and hole transport served as an upper bound. Rather, as demonstrated in the case of the ordered binary chain, we expect the activation energies which appear in the exponential variation of the conductance to be increased, and the mesoscopic structure to be changed.

The resistor network introduced in Sec. II is highly complex even in one dimension. The number of nodes increases exponentially with system size which limits any direct evaluation of the network conductance, by solving Kirchhoff's equations, for instance, to rather small systems. Furthermore the low-temperature regime, where correlation effects are expected to be most important, is difficult to reach because the individual conductances in the network become exponentially different in magnitude and this leads to numerical difficulties.

In such cases a percolative approach, such as the one first employed by AHL,[3] where one removes all resistors and then replaces them smallest first until the lattice percolates, has been shown to reproduce the exponential dependence of the resistance of the network quite well if the spread of the resistors is greater than about eight decades.[8,9] To what extent this bound is applicable to the many-particle networks is not clear since the distribution of resistors in the network is not homogeneous. Subnetworks of small and relatively larger resistors corresponding to low- and high-energy excited states span the network with the number of resistors in respective subnetworks increasing exponentially with the magnitude of the resistors. One can however convince oneself by examining the binary chain that the critical resistance at percolation still dominates the resistance at sufficiently low temperatures. Even if this condition is not satisfied this sort of approach is useful as it places a lower bound on the conductance.

Following AHL we remove all the resistors in our network and replace them, smallest first, up to some G_c, the smallest conductance at which the network first percolates. In the zero-temperature limit the conductance of the network will be dominated by G_c. The allowed conductances must satisfy the condition

$$G_{IJ} \geq G_c \tag{4.1}$$

or

$$G_c \leq \frac{P_I \gamma_{ij}}{kT} \ . \tag{4.2}$$

Considering for the moment those hops which lower the energy of the system, i.e., $E_I > E_J$, we may rewrite this in the form

$$1 \geq \frac{(E_I - \mu N_I + kT \ln Z)}{E_{\max}} + \frac{R_{ij}}{R_{\max}} \ , \tag{4.3}$$

where $E_{\max} = -kT \ln(G_c)$ and $R_{\max} = \ln(G_c)/\alpha$.

Thus instead of considering conductances one can consider adding nodes of increasing energy subject to the above constraint until the network percolates. We have not found any way of analytically solving this problem in the manner of AHL, so we have had to resort to a numerical investigation. Although the percolative approach simplifies the problem considerably, the exponential growth of the network with increasing system size limits the systems that can be examined, computer time being a more restrictive constraint then storage, to one-dimensional chains of up to 60 atoms. The most computationally efficient way of implementing the above procedure in one dimension that we have found is the following: One first locates the ground state, which must belong to the percolating cluster, and then explores allowed hops to the right. The allowed configurations are stored and their allowed right neighbors located. This procedure is repeated until a right hop leads to the ground state at which point the system has percolated, or until a dead end is reached where no more right hops are possible at which point $E_{\max}$ must be increased. The ground state can be located iteratively since the system is noninteracting, and the procedure can be started after all nodes with $E \leq E_{\rm AHL}$ have been added, as one knows that $E_{\max}$ must be greater then the mean-field result.

The results of such an investigation are presented below where we examine the effect of correlations on the percolation estimate of the conductance of random one-dimensional chains connected at either end to reservoirs which have the property of always being able to donate or accept particles if energetically favorable.

A. Numerical results for 1D chains

1. Nearest-neighbor hopping

The conductance of one-dimensional chains with nearest-neighbor hopping is known to be simply activated. At low temperatures the activation energy in mean-field theory is given by the largest energy barrier that needs to be overcome. The effect of taking correlations into account is shown in Fig. 3, which shows a plot of the fractional change in $E_{\max}$ due to correlations as a function of system size, averaged over an ensemble of systems. The site energies are uniformly distributed between 0 and W, with one barrier of height W to set the scale.

The change in activation energy $\delta E_{\max}$ increases with system size because the probability that a particle has to cross a pair of particle-hole barriers that dominate the activation energy increases. Closer examination shows that the particle-hole barriers have to be next to each other,

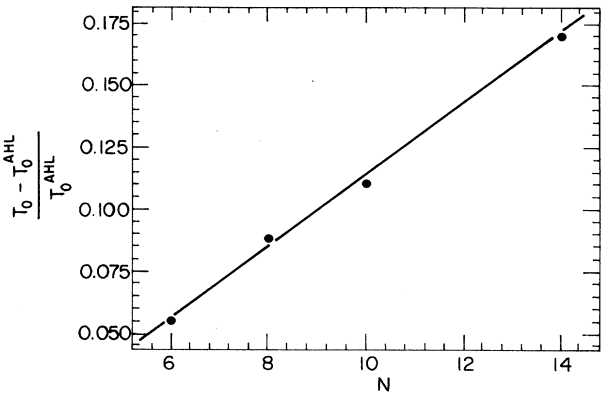

FIG. 3. The fractional change in the activation energy due to the inclusion of correlations as a function of the system size for a one-dimensional chain with nearest-neighbor hopping.

otherwise the system is able to make use of the reservoirs and smaller barriers to create room before crossing the largest. In addition the barriers have to be of roughly the same size. The four-site system with site energies 0, 1, $\frac{1}{2}$, and 1 illustrates this point.

For long chains it is highly probable that a pair of particle-hole barriers with energy difference W will appear, leading to the same activation energy and chemical potential variation as the ordered binary chain. The probability of such a configuration appearing increases linearly with system size which is consistent with Fig. 3. Correlation effects are thus always important in nearest-neighbor hopping.

2. Variable range hopping

In the Mott hopping regime the conductance of one-dimensional systems is known to behave as

$$\sigma(T) \sim \exp[-(T_0/T)^{1/2}] \ , \tag{4.4}$$

before crossing over to a simply activated form for sufficiently long chains.[10] T_0 depends upon the length of the chain L, and within mean-field theory is well approximated by[11]

$$T_0 \sim \alpha \ln(2\alpha L) \ . \tag{4.5}$$

In order to examine the role of correlations in the variable-range regime we have calculated the change in T_0 for various values of L and α, averaged over about 100 samples. The temperatures at which the calculations are performed have to be carefully chosen so that one is in the $T^{-1/2}$ regime. For short chains the temperature interval over which T_0, defined as $E_{\max}^2/T$, is temperature independent is small. At too low temperatures the carriers essentially hop right across the system, and T_0 increases with increasing temperature as the activation energy increases. At high temperatures the problem becomes nearest neighbor like and the activation energy is fixed so T_0 decreases with increasing temperature.

A plot of the change in T_0 due to correlations as a function of T_0 is shown in Fig. 4. The data are fairly

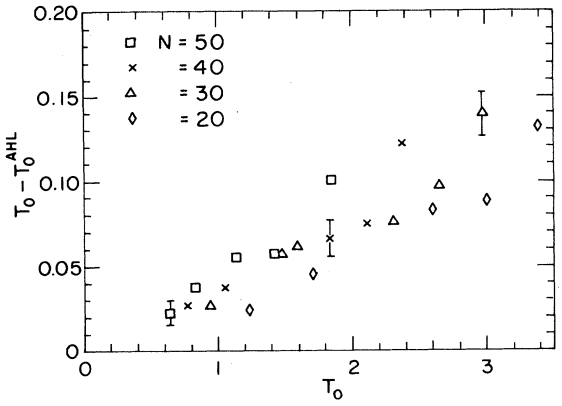

FIG. 4. The change in T_0 plotted as a function of T_0 for one-dimensional chains of various lengths with variable-range hopping. Some typical error bars are shown. Units are arbitrary.

scattered, because short chains have poor statistics,[12] and probably more so because the conditions mentioned above are difficult to implement in practice, but several trends can be noticed. δT_0 decreases with T_0 as would be expected, since the average energy barrier that has to be crossed decreases, and for a given value of T_0 the larger system sizes tend to show larger differences, especially at large values of T_0. This is probably because increasing L while keeping T_0 fixed decreases the average distance between hops.[11] The particles in the longer samples thus have to make, on average, a greater number of hops and hence the chances of correlations being important are increased.

The interesting regime is the low-T_0 regime since this corresponds to the thermodynamic limit, where one takes L to infinity keeping αL constant, but unfortunately our system sizes are too small to probe this region. A simple linear extrapolation from the available data yields changes of about 4% in T_0, but the effect may be larger given the tendency of δT_0 to increase with L. An upper limit for δT_0 of $3T_0$ can be obtained by noticing that $E_{\max}$ can at most be twice its mean-field value. One has

$$0 \leq \delta T_0 \leq 3T_0 \ . \tag{4.6}$$

The conductance behaves as

$$\sigma \sim \sigma_{\mathrm{AHL}}^{(1+\delta T_0/T_0)^{1/2}} , \tag{4.7}$$

so it is the value of $\delta T_0/T_0$ for small T_0 which is important.

Correlation effects are more striking if one examines the mesoscopic structure of a single sample. Recent experimental work on quasi-one-dimensional systems[6] has revealed large reproducible noiselike structure in the conductance with variation of the Fermi level. Lee[7] proposed that such structure was due to single hops controlling the resistance of the sample over a small range of values of the chemical potential and, by approximating the conductance of the chain by the critical conductance at percolation, using the mean-field networks, was able to show that mean-field theory could indeed reproduce much of this structure. Two types of hops were found to be important. When the dominant hop involved states on opposite sides of the Fermi level the conductance was found to be independent of the chemical potential, while if the states were both above or below the Fermi level a linear variation with slope $1/T$ was observed. The structure consisted of peaks some with flat tops, as the dominant hop changed with the chemical potential.

Experimentally the flat tops have never been observed, and it is interesting to speculate whether taking account of correlations may modify the flat tops as they do in the case of the ordered binary chain discussed in Sec. II where it as always cheaper to introduce an extra particle from the reservoir in order to lower the activation energy. Correlation effects are expected to modify this structure since it is not the dominant single hop but rather the largest energy barrier which needs to be crossed which controls the resistance at low temperatures.

Figure 5 shows a typical example where we have plotted both the mean-field and correlation resistance for a chain of 60 atoms with $\alpha L = 20$, so typically only a few hops are required to traverse the system as in the experimental situation.[6] Correlation effects remove the flat regions predicted by mean-field theory, and the results resemble experimental data much better than the mean-field curve but there are some features, such as the sharp valleys and round peaks, that we believe arise from the percolation approximation although some of these features seem to appear in the experimental data. The source of peaks and valleys can be deduced from Eq. (2.10). The critical conductance must have the form

$$\ln G_C \sim -\frac{(E_C - \mu N_C) - F}{kT} - \alpha R_{ij} \ , \tag{4.8}$$

where $F = -kT \ln Z$ is the free energy of the system.

At low temperatures the free energy of the system is well approximated by the ground-state energy

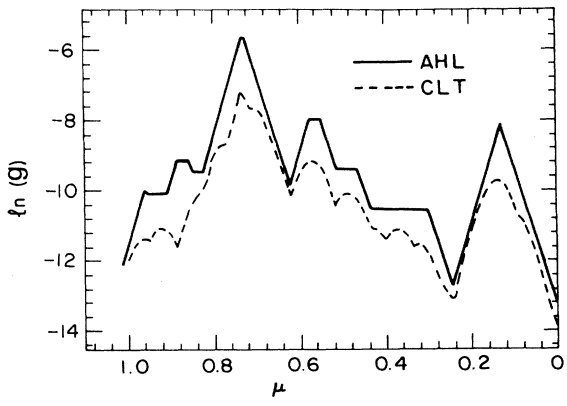

FIG. 5. A plot of the logarithm of the conductance $\ln(g)$ against μ, the chemical potential for a one-dimensional chain of 60 atoms, with $\alpha L = 20$. The solid curve in the mean-field prediction, and the dashed line the result of including correlations. Units are arbitrary.

$E_{g.s.} - \mu N_{g.s.}$ except when the system is about to change ground states, when contributions from two states become important. Ignoring this for the moment it can be seen that the structure of the peaks and valleys is related to whether the dominant hop contains more or less particles than the ground state. Large fluctuations in particle number are not favored so most of the peaks will have slopes of $1/T$ although larger slopes are also possible. Peaks correspond to the situation where the dominant hop goes from having more particles than in the ground state to having less, so the ground state must change across a peak. Across a valley the ground state is likely to remain fixed, the system simply goes from having one less particle to one more.

The rounding of the peaks is related to the change in ground state. Near the peak center the free energy is smaller than its zero-temperature value because contributions from the ground states on either side become important. The cross over between the two regions does not occur with a discontinuity as at zero temperature but smoothly with a nonlinear dependence on μ. Thus from Eq. (4.8) the reduced free energy leads to a smaller conductance. Valleys are not rounded because the ground state is likely to remain fixed. In reality not all peaks would be effected by this mechanism because if the dominant hop changes across a peak then two critical paths become important near the peak center and this offsets the decrease in the free energy. All the valleys would be rounded because across a valley the dominant hop always changes, while the free energy is fixed, and so the effect of two critical paths in parallel would be to increase the conductance compared with the percolation estimate.

These arguments do not rule out the possibility of having flat tops to peaks or bottoms to valleys, but we have not observed any so far. Interestingly if the free energy is replaced everywhere by its zero-temperature value, these do occur; so it appears that the free energy is important here as well. It may be that at the temperatures where variable-range hopping is operative flat regions are always rounded out. Special examples can be constructed where flat regions appear at sufficiently low temperatures.

Webb *et al.*[6] also followed the position of given peaks as a function of the temperature and found that some shifted to larger values of μ, some shifted to smaller values and some did not shift at all. This behavior is simple to understand within the many-particle formalism. Consider a given valley. The dominant hop changes across a valley and using Eq. (4.8) it easy to show that the position of the valley minimum has a temperature dependence of the form

$$\mu_p \sim \alpha(R_{12} - R_{34})T \ , \tag{4.9}$$

where R_{12} and R_{34} are the distances a particle hops in the dominant hop on either side of the peak. The sign of the shift thus depends upon the difference in hopping distances on either side of the valley. Thus all valleys should shift except those rare ones where there is no asymmetry in the hopping distances. Peaks, on the other hand, can frequently exhibit no shift since the dominant hop can remain fixed across a peak.

V. CONCLUSIONS

We find that correlation effects are important to a correct understanding of hopping conductivity. At low temperatures it is not the dominant single-particle hop which determines the conductance in the Mott hopping regime but the dominant hop between many-particle configurations. Correlation effects are found to lead to increased activation energies and modified mesoscopic behavior in the hopping conductivity of one-dimensional systems. While the amount by which the activation energy is increased is clear in the case of nearest-neighbor hopping, more work needs to be done on longer chains in the variable-range case, before something can be said about the thermodynamic limit. It may be possible, by making further approximations in the percolation procedure, to do this and also to study higher-dimensional systems. The inclusion of correlations leads to a mesoscopic behavior which is much closer to that experimentally observed in one dimension than that predicted by mean-field theory. The peaks and valleys are related to whether the dominant hop between many-particle configurations contains more or less particles than the ground state and not to whether the dominant single-particle hop connects states above or below the Fermi level, as in mean-field theory. Within the percolation approximation we find that there are clear differences between peaks and valleys. Across a peak the ground state changes and this rounds the peak, while-across a valley the ground state remains fixed and they remain sharp. In reality the valleys would be also be rounded due to the importance of two conducting paths near the valley minimum. No flat regions were observed as in the experiments.

In mean-field theory and in our calculations Coulomb interactions between carriers have been ignored. In systems where the states are strongly localized these are of long range as screening is not effective and are known to lead to the formation of a soft gap[13] in the density of states. It should be possible to understand the role of Coulomb interactions in one dimension by using the many-particle networks.

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