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Abstract
We have studied one aspect of the effect of Coulomb interactions on the hopping
conductivity of a band of localized electrons in a disordered system, that is
correlations in successive hops due to the Coulomb gap. At low temperatures,
the correlation holes around sites involved in the hop do not relax, and there
is an enhanced probability for backward hops. We calculate both dc and
ac conductivities by considering correlated random walks in the disordered
medium.

1. Introduction

In this paper we shall be concerned with the role of Coulomb interactions on the hopping
conduction of localized electrons at low temperatures. In a classic paper, Mott [1] showed
that as the temperature is lowered, the conductivity changes from its usual activated form
$\sigma_{dc} \propto \exp(-E_3/k_B T)$ to a form $\sigma_{dc} \propto \exp[-(T_M/T)^{1/4}]$. This has been termed 'variable range
hopping' (VRH), as the carrier tends to optimize between the distance of the hop and the energy
barrier of the hop. Mott's formula has been used quite successfully to analyse a large number
of experiments of hopping transport in impurity bands and amorphous semiconductors [2].
However, it was realized early by Efros and Shklovskii [3] that this picture ignores Coulomb
interactions between the carriers. Coulomb interactions change the temperature dependence
derived by Mott in a qualitative manner, i.e. to [3] $\sigma_{dc} \propto \exp[-(T_{ES}/T)^{1/2}]$. This is
understood in terms of the Coulomb gap, whose existence had been shown earlier by Pollak [4]
and Srinivasan [5]. They argued that in a system of localized electrons, the ground state
configuration of electron occupation is such that there would be a soft gap in its excitation
energy. This has been termed the 'Coulomb gap', and means that the single-particle density
of states (DOS) of excitation energy has the form $\rho(\epsilon) \propto (\epsilon - \epsilon_F)^2$ around the Fermi energy
ϵ_F. There have been numerous studies [6], in which the $T^{1/2}$ behaviour has been observed.
Also in several studies [7, 8] one sees a crossover from $T^{1/4}$ behaviour (Mott's regime) to $T^{1/2}$
behaviour (Coulomb gap regime) as temperature is lowered.

Mott's VRH arguments have been formalized using the rate equation approach of Miller
and Abrahams [9]. The linearized rate equation can be solved using standard Green function

techniques as applied to disordered systems. Physically, this amounts to calculating the conductivity of a random resistor network which is also equivalent to the calculation of diffusion of a single particle executing a random walk in a disordered medium. The main problem with such approaches lies in incorporating the effects of Coulomb interactions, as these are basically single-particle approximations of transport. The analysis of Efros and Shklovskii [3] incorporates Coulomb interactions only through the Coulomb gap in the DOS, which again allows the use of the single-particle picture.

There have been several efforts [10–13] to incorporate the effect of Coulomb interactions on hopping conduction. Building on the work of Dersch *et al* [13], Lamba and Kumar [14] have given a detailed many-body treatment to describe the crossover from Mott's to the Coulomb gap regime. This crossover has also been well described by some empirical approaches [7, 15]. In order to consider other aspects of correlations, it is important to keep in mind the physical nature of the Coulomb gap. The Coulomb gap is basically the difference between the energy of an occupied site and a unoccupied site around the Fermi level. This happens due to correlations in the occupation of sites of the following kind. In the neighbourhood of an occupied site, the occupation of other electrons is less than average, which in turn reduces the Hartree energy associated with the occupied sites near the Fermi level. Similarly the neighbourhoods of unoccupied sites have larger than average occupation of electrons (negative holes), thereby raising their Hartree energy.

Thus when an electron jumps from an occupied site to an unoccupied site, it has to overcome an energy barrier of the order of the Coulomb gap. However, this assumes that the electron hops with the other electrons frozen at their sites. If one allows for the relaxation of correlation clouds around the two sites between which the electron hops, then the energy barrier also relaxes. The electron hopping which occurs concomitant with the charge relaxation was termed 'polaron' hopping by Mott [16]. He further argued that under these conditions the $T^{1/4}$ law should apply. Using an explicit model, Lamba and Kumar [14] argued that the relaxation of charge clouds responsible for the Coulomb gap is related to the conductivity itself. So at low temperatures when the relaxation is low due to small conductivity, the charge hopping is occurring in the frozen background of charge, and the Coulomb gap effects the conduction process. With increasing temperature, the Coulomb gap relaxes [14], leading to a crossover from the $T^{1/2}$ to the $T^{1/4}$ law.

The purpose of the present paper is to deal with one other aspect of correlated motion of carriers in the calculation of the dc and ac conductivities in the Coulomb gap regime. In particular, we wish to take into account backward correlations in hopping, which have been observed in numerical simulations of conductivities. In particular, in the simulations of Maass *et al* [17], it is seen that when disorder and Coulomb interactions are of comparable strength, the successive carrier hops are strongly correlated. A carrier is likely to jump back to the site from which it came with greater probability than in uncorrelated diffusion. This can be physically understood in the terms discussed above. At low temperatures in the $T^{1/2}$ regime, the relaxation of holes as the carrier jumps is slow, so there should be a tendency for the electron to go back to the hole it left behind in the previous jump, as the other unoccupied sites have energies higher by the order of the Coulomb gap.

In this paper, we consider the effect of this correlation using a random walk model and relate the enhanced backward hopping rate to the Coulomb gap by a simple consideration. Correlated random walks [18, 19] are a class of random walks in which, unlike Markovian walks, the memory is not lost after each step. Correlated random walks have various physical applications. The two most important are conformation of polymers and tracer diffusion in metals. Most of the theoretical work has concentrated on correlated random walks on ordered lattices. Hilfer and Orbach [20] and Hilfer [21] have introduced disorder into this formalism by

considering walks on a bond percolation model. We extend their treatment to a more general class of disordered systems. Our treatment is restricted to the memory of one last step only.

2. Correlated random walk

Consider the hopping of a single particle in a disordered medium. For a Markovian random walk, we start with writing a master equation for $P(i, t)$, which is the conditional probability to find a particle at site i at time t if it started from the origin at time $t = 0$. The effect of correlation is to retain the memory of the last step. So one defines the probability $P(i, j, t)$ to find a walker at site i at time t given that it arrived at i via a direct transition from site j. The probability density $P(i, t)$ is obtained by summation over all possible histories in $P(i, j, t)$ as

$$P(i, t) = \sum_j P(i, j, t), \tag{1}$$

where the sum is over all sites of the system excluding i. The initial condition that the walker is at the origin (o) at time $t = 0$ is $P(i, 0) = \delta_{i,o}$. A transition rate W_{ij}^{b} is assigned to a jump from site j to site i if the previous jump was from site i to site j. A transition rate W_{ij} is assigned to a jump from site j to i if the particle had jumped to j from any site except i. The conditional probability $P(i, j, t)$ obeys the following master equation [19, 20]

$$\dot{P}(i, j, t) = \sum_{k \neq i} W_{ij} P(j, k, t) + W_{ij}^{\mathrm{b}} P(j, i, t) - \left[\sum_{k \neq j} W_{ki} + W_{ji}^{\mathrm{b}} \right] P(i, j, t), \tag{2}$$

where $\dot{P}$ implies a time derivative of P. If we define $\delta W_{ij} = W_{ij}^{\mathrm{b}} - W_{ij}$, equation (2) can be rewritten as

$$\dot{P}(i, j, t) = W_{ij} P(j, t) + \delta W_{ij} P(j, i, t) - \left[\sum_k W_{ki} + \delta W_{ji} \right] P(i, j, t). \tag{3}$$

Summing over j in equation (3) we get an equation for $P(i, t)$

$$\dot{P}(i, t) = \sum_j W_{ij} [P(j, t) - P(i, t)] + \sum_j \delta W_{ji} A(j, i, t), \tag{4}$$

where we have defined

$$A(i, j, t) = P(j, i, t) - P(i, j, t). \tag{5}$$

Using equation (3) we can write an equation for $P(j, i, t)$ and thereby obtain an equation for $A(i, j, t)$. One can get a closed set of equations, by replacing $\sum_k W_{ki}$ by W_{t} which is its average over sites, $W_{\mathrm{t}} = \langle \sum_k W_{ki} \rangle$. This yields the equation

$$\dot{A}(i, j, t) = W_{ij} [P(i, t) - P(j, t)] - [W_{\mathrm{t}} + 2 \delta W_{ij}] A(j, i, t). \tag{6}$$

The approximation made here seems physically reasonable for the two-site probabilities and avoids introduction of higher-order multi-site probabilities. Using initial conditions $P(i, 0) = \delta_{i,o}$ and $A(j, i, 0) = 0$ and taking Laplace transform of equations (4) and (6), we obtain

$$s\bar{P}(i, s) - P(i, 0) = \sum_j [\bar{P}(j, s) - \bar{P}(i, s)] \left[W_{ij} - \frac{\delta W_{ij} W_{ij}}{s + W_{\mathrm{t}} + 2 \delta W_{ij}} \right], \tag{7}$$

where $\bar{P}(i, s)$ denotes the Laplace transform of $P(i, t)$. We now make a further assumption by taking δW_{ij} to be site-independent, as we later relate it to the Coulomb gap. Replacing δW_{ij}

by its average value δW_b, we get an equation equivalent to the Markovian rate equation, but with a scaled frequency $\tilde{s} = s h(s)$ where

$$h(s) = \left[1 + \frac{\delta W_b}{s + W_t + \delta W_b} \right]. \tag{8}$$

Equation (7) is solved in terms of the Green function $G_{ij}(s)$

$$P(i, s) = h(s) \sum_j G_{ij}(\tilde{s}) P(j, 0), \tag{9}$$

where in the matrix notation $\hat{G}$ obeys the equation

$$\hat{G}(\tilde{s} - \hat{W}) = I, \tag{10}$$

with

$$\hat{W}_{ij} = W_{ij} - \delta_{ij} \sum_k W_{ki}. \tag{11}$$

3. Model

We consider a system of randomly placed sites with a density n. Each site has an energy ϕ_i, where ϕ_i are randomly drawn from a rectangular distribution taken to be

$$P(\phi) = \frac{1}{2W} \qquad |\phi| \leqslant W, \tag{12}$$

$$P(\phi) = 0 \qquad |\phi| > W. \tag{13}$$

The DOS for the site energies has the form

$$g(\phi) = g_o = \frac{n}{2W} \qquad |\phi| \leqslant W. \tag{14}$$

In the presence of Coulomb interactions, the single site energies are modified by Hartree terms and take the form

$$\epsilon_i = \phi_i + \sum_j K_{ij} n_j, \tag{15}$$

where $K_{ij} = e^2 / \kappa R_{ij}$ denotes the Coulomb interaction between electrons at the sites i and j, n_j are the site occupation numbers and κ is the dielectric constant of the medium. As shown by Efros and Shklovskii [3], the stability of the ground state requires that the DOS $g(\epsilon)$ for the Hartree energies has a gap around the Fermi level ϵ_F, and is given by

$$g(\epsilon) = \frac{C(\epsilon - \epsilon_F)^2 \kappa^3}{e^6} \qquad |\epsilon - \epsilon_F| \leqslant \Delta, \tag{16}$$

$$g(\epsilon) = g_o \qquad \Delta < |\epsilon - \epsilon_F| < W, \tag{17}$$

where C is a dimensionless constant of order unity and Δ is the width of the Coulomb gap, which is obtained by continuity to be

$$\Delta = \frac{e^3 g_o^{1/2}}{\kappa^{3/2}}. \tag{18}$$

Next we model δW_b. It is the difference between the probability of a jump between sites around the Fermi level and the probability of the backward hop. For temperatures $k_B T \ll \Delta$, we assume that most of the unoccupied sites available to the electron have energies higher by order Δ and the backward hop is a downward jump in energy of this order, so we take

$$\delta W_b = W_t \tanh \left(\frac{\Delta}{k_B T} \right). \tag{19}$$

For temperatures, $k_B T \ll \Delta$, $\delta W_b \approx W_t$, which makes backward hops as probable as the jump probability to all other sites. As the temperature increases, this quantity decreases as expected. In this paper we have focused on the low-temperature regime.

The general transition rates in the Coulomb glass problem are the asymmetric rates given by, $T_{ij} = \nu_0 \exp\left[-\left(\frac{R_{ij}}{a} + \frac{|\epsilon_{ij}| + |\epsilon_i| + |\epsilon_j|}{k_B T}\right)\right]$ where ϵ_i, ϵ_j are Hartree energies of the sites i and j defined in equation (15), $\epsilon_{ij} = \epsilon_i - \epsilon_j - K_{ij}$ and ν_0 is taken to be the phonon Debye frequency. At low frequencies where $\sigma(\omega) = \sigma(0)$ (see the results section), the VRH and percolation pictures tell us that the critical hop is a long hop. As the frequency increases and dispersion in conductivity sets in, the hop distance decreases and is approximately proportional to $\ln(\omega)$. So at high frequencies the K_{ij} factor becomes important. We have neglected the K_{ij} factor in our calculation, so our results are not valid at high frequencies. In earlier calculations on dc conductivity, we have numerically checked [14] that the conductivity results do not change qualitatively if we replace the asymmetric transition rates by the symmetric transition rates, $W(R, \epsilon) = \nu_0 \exp\left[-\left(\frac{R}{a} + \frac{\epsilon}{k_B T}\right)\right]$. From this, for $k_B T \ll \Delta$, we obtain

$$W_t = 16\pi (k_B T)^3 a^3 n \kappa^3 / e^6. \tag{20}$$

We work in the frequency units of ν_0.

4. Calculation of conductivity

For the calculation of conductivity in a disordered system, several effective medium approximations have been proposed [22]. All of them require a self-consistent calculation. In the present calculation, we adopt the procedure of Movaghar and Schirmacher [23]. In this approach one makes an effective medium approximation using the renormalized perturbation expansion. This procedure has given fairly good results for conductivity in disordered systems [24, 25], however, it is difficult to implement this method in the presence of energy disorder and asymmetric transition rates. So in this paper we use the symmetric transition rates mentioned above. The expression for frequency-dependent conductivity can be written as

$$\sigma(\omega) = \frac{(i\omega)^2 e^2 h(\omega)}{6 V k_B T} \sum_{ij} \langle F(\epsilon_i) R_{ij}^2 G_{ij}(\tilde{\omega}) \rangle, \tag{21}$$

where the angular bracket denotes the configurational average, $i\tilde{\omega} = \tilde{s}$ is the scaled frequency defined by equation (8), and $F(\epsilon_i) = f(\epsilon_i)(1 - f(\epsilon_i))$, where $f(\epsilon_i)$ is the Fermi function. Since our transition rates are symmetric, we take $F_i = F = q(1 - q)$ for all i, where q is the number of electrons per site. Using a two-site approximation for solving equation (10), an effective transition rate $g_{ij}(\omega)$ has been derived in [23], which obeys the equation

$$g_{ij} = \left[\frac{1}{W_{ij}} + \frac{1}{i\tilde{\omega} + \sum_l g_{jl}}\right]^{-1}. \tag{22}$$

The self-consistent equation for the average value $\sigma_1(\omega)$ of $\sum_j g_{ij}$ can be recast into the form

$$\sigma_1(\tilde{\omega}) = (\sigma_1(\tilde{\omega}) + i\tilde{\omega}) \int \frac{g(\epsilon) W(R, \epsilon) \, d^3 R \, d\epsilon}{(\sigma_1(\tilde{\omega}) + i\tilde{\omega}) W(R, \epsilon) + 1}, \tag{23}$$

and the conductivity is given as

$$\sigma(\omega) = \frac{F e^2}{6 V k_B T h(\omega)} \langle R^2(\tilde{\omega}) \rangle \sigma_1(\tilde{\omega}), \tag{24}$$

with

$$\langle R^2(\tilde{\omega}) \rangle = \left\langle \frac{R^2 W(R, \epsilon)}{\sigma_1(\tilde{\omega}) + i\tilde{\omega} + W(R, \epsilon)} \right\rangle \bigg/ \left\langle \frac{W(R, \epsilon)}{\sigma_1(\tilde{\omega}) + i\tilde{\omega} + W(R, \epsilon)} \right\rangle. \tag{25}$$

Using transition rates defined in section 3 and the condition $\Delta \gg k_B T$, equation (23) can be written in terms of dimensionless variables as

$$\sigma_1(\tilde{\omega}) = (\sigma_1(\tilde{\omega}) + i\tilde{\omega})\, p \int\int \frac{\epsilon^2 r^2 \, dr \, d\epsilon}{(\sigma_1(\tilde{\omega}) + i\tilde{\omega})W(r,\epsilon)^{-1} + 1} \tag{26}$$

where $p = 4\pi n a^3 (k_B T)^3 \kappa^3 / e^6 = W_t/4$. Equations (24) and (26) differ from the earlier work in two respects. First, the frequency ω is replaced by the scaled frequency $\tilde{\omega}$, given by equation (8). Secondly, there is a prefactor of $h(\omega)$ in the expression of conductivity.

5. Results

We first look at the calculation of conductivity for the uncorrelated system. The previous analysis [24, 25] was done by neglecting the frequency dependence of mean square displacement, which is rather weak. Thus the frequency dependence of conductivity is largely determined by $\sigma_1(\omega)$. Analysis of equation (26) shows that the significant dispersion in $\sigma_1(\omega)$ sets in only when frequency becomes greater than $\sigma_1(0)$. The equation for $\sigma_1(0)$ is equivalent to the percolation criterion employed by Ambegaokar *et al* [26]. Solving equation (26) for $\omega = 0$ one gets

$$\sigma_1(0) = \exp(-u) = \exp\left[-\left(\frac{180}{p}\right)^{1/6}\right] = \exp\left[-\left(\frac{T_{es}}{T}\right)^{\frac{1}{2}}\right]. \tag{27}$$

In the infinite frequency limit, $\sigma_1(\omega)$ is proportional to the total transition rate W_t. In the intermediate frequency range the conductivity obeys a power law in frequency, $\sigma_1(\omega) \propto \omega^s$. The slope is crudely given by $s = (u + \ln W_t)/u$. The slope increases as the temperature p decreases.

For the correlated walk, the conductivity becomes a function of a scaled frequency $\tilde{\omega}$ instead of ω. The behaviour of σ_1 as a function of ω and $\tilde{\omega}$ is shown in figures 1 and 2 for two different temperatures. The curve for $\sigma_1(\tilde{\omega})$ shifts to the left with respect to $\sigma_1(\omega)$ on the frequency scale. The shift depends upon the frequency-dependent correlation factor $h(\omega)$. As seen from figures 1 and 2, the shift is apparent for the frequency range $\sigma_1(0) < \omega < W_t$. At high frequencies, $\omega \gg W_t$, $h(\omega) \approx 1$ so the correlation effect is negligible. Since there is no dispersion in the low-frequency range $\omega < \sigma_1(0)$, the curves again coincide. The crossover frequency W_t is temperature-dependent and does not scale with $\sigma_1(0)$. Comparison of figures 1 and 2 shows that the crossover from correlated to uncorrelated hopping occurs at different frequencies for different temperatures.
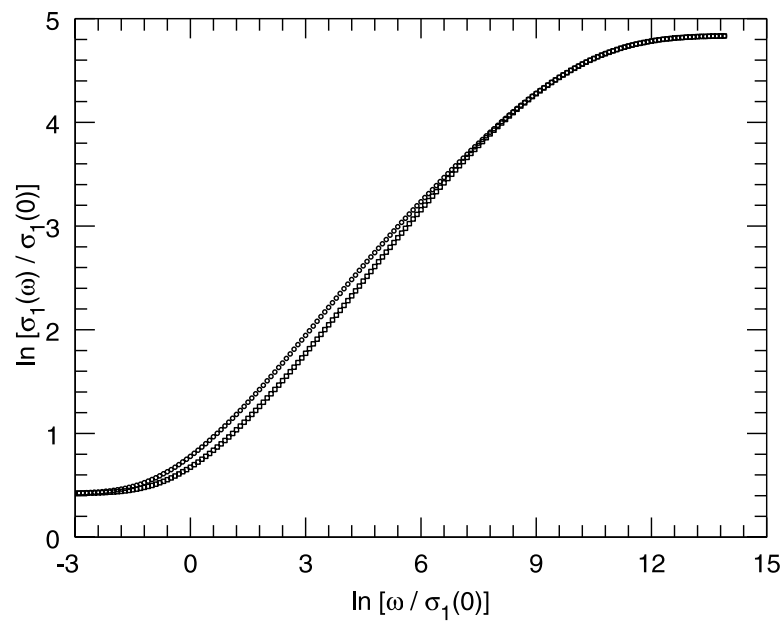
The calculation of conductivity with correlated hopping requires computation of the factor $\langle R^2(\tilde{\omega})\rangle$. This leads to the reduction of the conductivity by a factor of $1/h(\omega)$. The results for conductivity for two values of p are shown in figures 3 and 4. At temperatures reported here, $h(0)$ is weakly dependent on temperature, so the change in dc conductivity is also weakly dependent on p, as seen in these figures. At higher frequencies $\omega > W_t$, $h(\omega) \approx 1$, so no correlation effects are present and the curves coincide. Enhanced backward correlations increase the slope of conductivity by $\ln[h(0)]/u$. This is significant at high temperatures (small u). The increase in slope due to backward correlations decreases as temperature decreases (u increases), as seen in figures 3 and 4.

We next discuss the scaling of the ac conductivity, which has been seen in numerous studies [27, 28]. The experimental studies show that over a considerable range of temperatures scaling of the form

$$\sigma(\omega)/\sigma_{dc} = \Psi(\omega/T\sigma_{dc}), \tag{28}$$
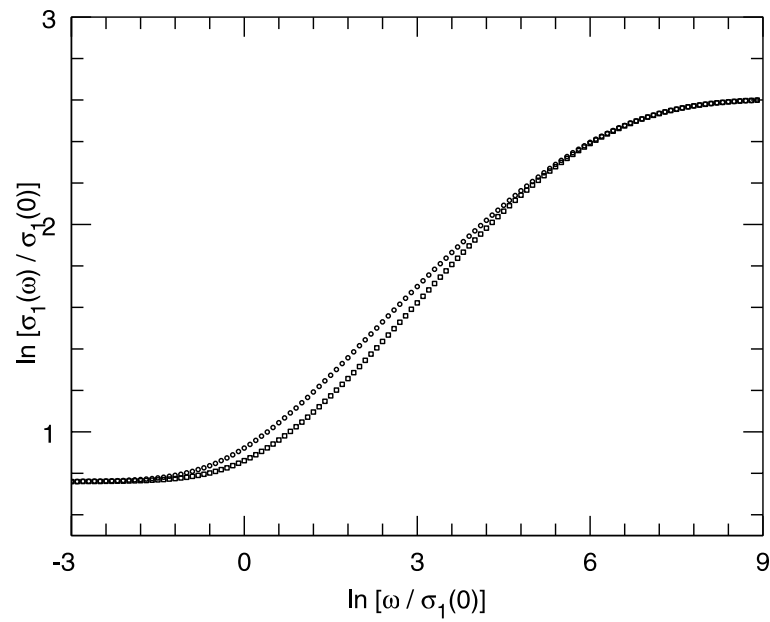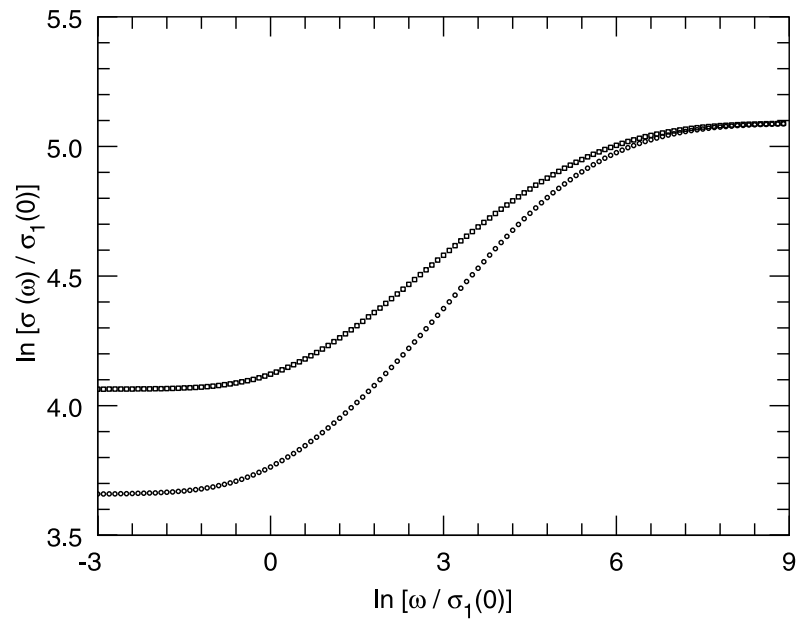
Figure 1. Plots of $\ln[\sigma_1(\omega)/\sigma_1(0)]$ with $\ln[\omega/\sigma_1(0)]$ for (O) correlated and (□) uncorrelated hopping for $p = 0.0002$.
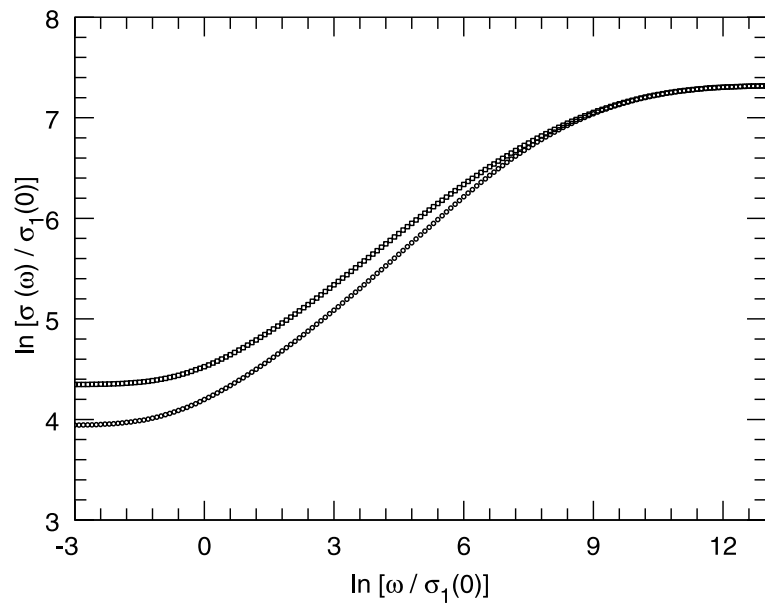

Figure 2. Plots of $\ln[\sigma_1(\omega)/\sigma_1(0)]$ with $\ln[\omega/\sigma_1(0)]$ for (O) correlated and (□) uncorrelated hopping for $p = 0.00002$.

holds for a wide range of materials, including ionic conductors. In the present calculation we have already seen that the frequency dispersion of conductivity sets in only when

Figure 3. Logarithmic plots of scaled conductivity $\ln[\sigma(\omega)/\sigma_1(0)]$ against scaled frequency $\ln[\omega/\sigma_1(0)]$ for (O) correlated and (□) uncorrelated hopping for $p = 0.0002$.

Figure 4. Logarithmic plots of scaled conductivity $\ln[\sigma(\omega)/\sigma_1(0)]$ against scaled frequency $\ln[\omega/\sigma_1(0)]$ for (O) correlated and (□) uncorrelated hopping for $p = 0.00002$.

$\omega > \sigma_1(0) \propto T\sigma_{dc}$. The reason for this is quite apparent in equation (23). However, the scaling of equation (28) requires that $\sigma(\omega)/\sigma_{dc}$ will not have any temperature dependence apart from that contained in $\sigma_1(0)$. This, as in earlier studies [27], is true when the temperatures are

low and a Sommerfeld approximation can be made to evaluate the integrals in equations (23) and (25). Since $\tilde{\omega} = 0$ for $\omega = 0$ the equation for $\sigma_1(0)$ is the same for both correlated as well as Markovian hopping. However, for the correlated hopping the difference comes in the evaluation of the dc conductivity in which there is an additional factor of $h(0)$. Thus we can write

$$\sigma(\omega)/\sigma_{dc} = \Psi\left(\frac{\tilde{\omega}}{\sigma_1(0)}\right) = \Psi\left(\frac{\omega h(\sigma_1(0))}{\sigma_{dc} T h(0)}\right). \tag{29}$$

This shows that the scaling frequency changes due to correlated diffusion by the factor $h(\sigma_1(0)/h(0))$, which has been termed as the Haven ratio [28]. This factor has been introduced in an earlier work on ionic conduction on an empirical basis. Our work provides a quantitative estimate of the Haven ratio in the Coulomb gap regime.

6. Concluding remarks

In this paper, we have treated the effect of Coulomb interactions on hopping transport from two aspects. These are:

(a) depletion of single-particle DOS, which has a dominant effect on the temperature dependence of dc conductivity,
(b) the enhanced backward hopping, which is a consequence of an unrelaxed Coulomb hole (anti-hole) around an occupied (unoccupied) site.

The latter aspect has a considerable influence on the ac conductivity in the dispersive regime. We show that the scaling property of conductivity with frequency is still obeyed approximately. We also provide a quantitative estimate of Haven's ratio in the Coulomb gap regime of conduction.

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