Accurate DC Conductivity for Hopping Conduction within the Continuous Time Random Walk Approach†

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The continuous time random walk approach for hopping conduction in disordered systems is modified to obtain accurate DC conductivity values over a wide range of hopping site densities. The modification involves eliminating a small fraction of particularly isolated hopping sites from the determination of the waiting-time distribution for hopping. The elimination of these sites does not correspond to a pure truncation in the transition rate spectrum, but rather a truncation only in the distribution of contributing near-neighbor hopping distances. For a cutoff distance corresponding to the percolation radius r_p, the DC conductivity is within about 1 order of magnitude of percolation-based models and simulation results over a moderate range of hopping site densities. For a slightly larger cutoff distance of $1.15r_p$, the DC conductivity is in excellent agreement with the percolation-based theory over a wide range of hopping densities. The introduction of the near-neighbor cutoff does not influence the high-frequency conductivity, where the full CTRW provides an excellent description of the conductivity. Hence, the present simple modification permits an accurate and full interrogation of the frequency-dependent conductivity within a single conceptual framework.

1. Introduction

The understanding and accurate prediction of transport properties of disordered materials has been an area of considerable interest and study over the past forty years. The problem of hopping conduction, wherein charge carriers or other excitations perform discrete transitions among spatially and/or energetically well-separated sites, has been of particular interest due to its application to doped semiconductors and spectral diffusion, for instance. However, a full description of charge or excitation transport over the full range of frequencies and disorder has not been developed. The continuous time random walk (CTRW) approach to AC conductivity, pioneered in seminal papers by Scher and Lax,[1] is formally exact at high frequencies and has been shown to yield excellent predictions at moderate frequencies and moderate to high densities of sites.[2] However, the prediction of the DC conductivity within the CTRW is rather poor for very low site densities because of the assumed uncorrelated nature of the hopping within the model. In contrast, models based on percolation concepts, wherein the transport is largely confined to a highly correlated path of sites, have led to excellent predictions for the DC conductivity but cannot be extended to higher frequencies.[3−5] Methods for extending exact high-frequency results to lower frequencies through the introduction of pair-[6] and higher-order cluster correlations[7,8] cannot be carried through to the DC limit. Ad-hoc approaches, such as combining the high-frequency CTRW and DC percolation results, yield satisfactory results over all frequencies but do not provide any insight into the physics of the frequency-dependent conductivity.[2]

In this paper, we show that a simple modification to the CTRW approach can lead to an accurate DC conductivity while maintaining the excellent high-frequency predictions of the original CTRW model. Within the context of the iso-energetic hopping problem, the modification involves eliminating from consideration those sites that are spatially isolated from all other sites in the random material. Here, "spatially isolated" means those sites with a nearest-neighbor site beyond some cutoff distance r_c. For sufficiently large r_c, the number of eliminated sites is quite small. Figure 1 provides a schematic of the allowed transitions in such a system. If initially populated by a carrier, hopping off of the spatially isolated sites will be prohibitively long such that they do not contribute appreciably to the DC conductivity. Similarly, it is unlikely that such sites will receive a carrier during transport since the minimum transition rate into such sites is also quite long. The irrelevance of such sites to the conductivity has been widely recognized within the percolation model[3−5,9] and has been a tenet of various hopping

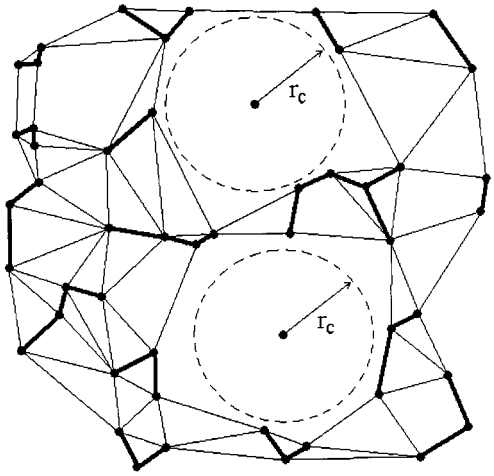

Figure 1. Schematic of hopping transport in a 2D spatially disordered system. Within the modified CTRW, isolated sites with near-neighbors beyond some cutoff radius r_c are excluded from consideration, while all transitions among all other sites are permitted. Heavy lines indicate near-neighbor transitions; light lines indicate some further-neighbor transitions; isolated sites are those with no near-neighbors within a cutoff radius.

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models.[6,10] The use of a cutoff in all of the transition rates as a means of obtaining percolation-like behavior was anticipated by Scher and Lax but was not investigated in detail.[1] The percolation model naturally suggests the cutoff distance to be $r_c = r_p$, where r_p is the critical distance between sites within the percolation model. Analysis below shows such a choice to provide the same scaling exponent of conductivity with density as found in the percolation model, but with an absolute magnitude difference in conductivity of about a factor of 10. An increased cutoff distance of $1.15r_p$ yields excellent results over a very wide range of densities. It should be noted that the present model for the DC conductivity is not as accurate as the analytic model derived by Butcher et al.[3,4] following on the Miller−Abrahams theory[11] and percolation concepts.[9] However, the present model does meld naturally with the CTRW model and hence to a highly accurate AC conductivity model.

The remainder of this paper is organized as follows. In the next section, we define the iso-energetic hopping model and its associated master equation formulation and present the basic CTRW approach to the conductivity. In section 3, we solve for the CTRW waiting-time distribution within the proposed modification and develop a near-analytic form for the DC conductivity. In section 4, results for the DC conductivity are presented and compared with the analytic model of Butcher et al.[3,4] In section 5, some physical implications of the modified CTRW are discussed.

2. CTRW Model for Hopping Conductivity

We consider a random configuration of sites at spatial positions $\{r_n\}$. Transport of charge between sites n and n' occurs with some transition rate $W_{n,n'}$, which is anticipated to depend primarily on the radial separation between the two sites. For a carrier located at site n_0 at time $t = 0$, the probability $P_n(t)$ that it is located at site n at time t is assumed to obey the master equation

$$\frac{dP_n(t)}{dt} = \sum_{n' \neq n} W_{nn'}P_{n'}(t) - \Gamma_n P_n(t) \tag{1}$$

where $\Gamma_n = \sum_{n' \neq n} W_{n'n}$ and the initial conditions are $P_n(0) = \delta_{n,n_0}$. An exact solution to the coupled eqs 1 is not feasible for a large number of sites. In addition, we wish to understand the behavior averaged over all possible configurations of sites.

To make analytic progress, Scher and Lax considered the associated problem of a continuous time random walk on a regular lattice wherein the transition time from lattice site to lattice site reflects the spectrum of transition times inherent in the true random system.[1] The quantity of interest in the CTRW is the waiting-time distribution $\psi(t)$, with $\psi(t)dt$ being the probability that the carrier will leave a site at time t given that it has been at that site since time $t = 0$. The relationship between $P_{n_0}(t)$ and $\psi(t)$ is then simply

$$\psi(t) = -\frac{d}{dt}\langle P_{n_0}(t)\rangle \tag{2}$$

where the brackets denote an averaging over the configuration of site positions in the system.

Within the CTRW, the AC conductivity for a density of n carriers of charge e at temperature T is given as

$$\sigma(\omega) = -\frac{ne^2}{6kT}\omega^2\sum_n (n - n_0)^2 P_n(\omega) \tag{3}$$

where $P_n(\omega)$ is the time Fourier transform of $P_n(t)$. In the DC limit of particular interest here, the conductivity becomes

$$\sigma_{DC} = \sigma(0) = \frac{ne^2}{kT}\frac{\sigma_{rms}^2}{6\bar{t}}$$

$$\sigma_{rms}^2 = \int_0^\infty dr r^2 g(r)$$

$$\bar{t} = \int_0^\infty dt t\psi(t) \tag{4}$$

where σ_{rms}^2 is the mean-square single hop distance, generally approximated as the mean square site separation, $g(r)$ is the near-neighbor radial distribution function (Hertz distribution) in the material, and $\bar{t}$ is the mean waiting time. Following from eq 2 and integration by parts,

$$\bar{t} = \int_0^\infty dt\langle P_{n_0}(t)\rangle \tag{5}$$

The DC conductivity thus follows directly from the site occupation probability $\langle P_{n_0}(t)\rangle$.

3. Modified CTRW for DC Conductivity

From the above discussion, the DC conductivity is seen to follow from the calculation of the average probability of finding the carrier on the initial site without having yet hopped away. This probability is obtained from the master eq 1 by eliminating the "supply" term $\sum_{n' = n} W_{nn'}P_{n'}(t)$ on the right hand side; since the carrier has yet to hop away, the initial site cannot be supplied by the carrier hopping back onto the initial site. For the initial site n_0, which we will now label as $n_0 = 1$, eq 1 then reads

$$\frac{dP_1(t)}{dt} = -\Gamma_1 P_1(t) \tag{6}$$

and hence

$$P_1(t) = e^{-\Gamma_1 t} \tag{7}$$

Equation 7 applies to any particular configuration, and includes the possible hopping from site 1 to all other sites $n = 2$, 3, etc. We must now average over the spatial locations of the surrounding sites, which are contained in the quantity Γ_1.

Here, we first specifically separate the nearest neighbor site, labeled 2, and average over the remaining sites $n = 3$, 4, etc. We will then subsequently average over the possible positions of the nearest neighbor site 2. Averaging over sites $n = 3$, 4, etc. yields

$$\langle P_1(t)\rangle_{3,4,etc.} = e^{-W_{12}t}\int_{r_2}^\infty \frac{d\vec{r}_3}{V}\frac{d\vec{r}_4}{V}...e^{-W_{13}t}\,e^{-W_{14}t}... = e^{-W_{12}t}\left(\frac{1}{V}\int_{r_2}^\infty d\vec{r}\,e^{-W_{1r}t}\right)^{N-2} \tag{8}$$

Adding and subtracting unity to the term in parentheses in eq 8, using the definition of the exponential, and defining the site density as $n_s = N/V$ leads to

$$\langle P_1(t)\rangle = e^{-W_{12}t}\,e^{-n_s\int_{r_2}^\infty dr' 4\pi r'^2(1 - \exp\{-W_{1r'}t\})} \tag{9}$$

where we have assumed W_{1r}, defined as the hopping rate between site 1 and a site at distance r, is a function only of the radial distance r. *We now average over the positions of the near-neighbor site 2, but eliminate those near-neighbor sites further than a cutoff distance* r_c. Taking site 1 to be at the origin $\vec{r} =$

0 and eliminating the subscripts for the subsequent sites, we obtain

$$\langle P_1(t)\rangle = \int_0^{r_c} \mathrm{d}r g(r)\, \mathrm{e}^{-W_r t}\, \mathrm{e}^{-n_s \int_r^\infty \mathrm{d}r' 4\pi r'^2 (1-\exp\{-W_{1r'}t\})} \quad (10)$$

The introduction of a cutoff near-neighbor separation r_c is the only modification we make to the original CTRW model. Note that this is a cutoff only for the near-neighbor distance; the possibility of parallel channels of carrier hopping via the further neighbors is still contained in eq 10. However, as the site density decreases and the site separations increase, transitions to further neighbors do become increasingly less likely, which is the physical basis of the percolation models.

To obtain numerical results and interrogate the effect of the choice of r_c on the conductivity, we now specialize to the isoenergetic hopping model with spatially random sites, for which

$$W_r = g_a \mathrm{e}^{-2\alpha r} \quad (11)$$

and

$$g(r) = 4\pi n_s r^2\, \mathrm{e}^{(4\pi/3) n_s r^3} \quad (12)$$

Our notation is chosen in accord with the numerical work of Butcher et al., to which our predictions will be compared.[3,4] Substituting eqs 11 and 12 into eq 10, using the dimensionless variables $\tilde{r} = 2\alpha r$ and $\tilde{t} = g_a t$, and defining $\tilde{x} = \ln(\tilde{t})$ and $\eta = 4\pi n_s/(2\alpha)^3$, leads to

$$\langle P_1(t)\rangle = \eta \int_0^{\tilde{r}_c} \mathrm{d}\tilde{r} \tilde{r}^2\, \mathrm{e}^{-\eta\tilde{r}^3/3}\, \mathrm{e}^{-\mathrm{e}^{\tilde{x}-\tilde{r}}}\, \mathrm{e}^{-\eta\int_{\tilde{r}}^\infty \mathrm{d}\tilde{r}'\tilde{r}'^2(1-\exp\{-\mathrm{e}^{\tilde{x}-\tilde{r}'}\})} \quad (13)$$

The calculation of $\bar{t}$ follows from eq 5 as

$$g_a \bar{t} = \int_{-\infty}^{\infty} \mathrm{d}\tilde{x}\, \mathrm{e}^{\tilde{x}} \eta \int_0^{\tilde{r}_c} \mathrm{d}\tilde{r}\tilde{r}^2\, \mathrm{e}^{-\eta\tilde{r}^3/3}\, \mathrm{e}^{-\mathrm{e}^{\tilde{x}-\tilde{r}}}\, \mathrm{e}^{-\eta\int_{\tilde{r}}^\infty \mathrm{d}\tilde{r}'\tilde{r}'^2(1-\exp\{-\mathrm{e}^{\tilde{x}-\tilde{r}'}\})} \quad (14)$$

With $g_a\bar{t}$ determined by eq 14, the DC conductivity can now be evaluated. The mean-square hopping distance can be estimated as, neglecting the cutoff radius so as to obtain analytic results,

$$\sigma_{\mathrm{rms}}^2 = \int_0^\infty \mathrm{d}r r^2\, 4\pi r^2 n_s\, \mathrm{e}^{(-4\pi/3) n_s r^3} = 3^{2/3}\Gamma\left(\frac{5}{3}\right)\frac{1}{4\alpha^2\eta^{2/3}} \quad (15)$$

The normalized DC conductivity then follows from eqs 4 and 15 as

$$\sigma_{\mathrm{DC}} = \frac{\sigma_{\mathrm{DC}}}{\left(\frac{ne^2 g_a \alpha}{n_s}\right)} = \frac{1}{24}\left(\frac{6}{\pi}\right)^{2/3}\Gamma\left(\frac{5}{3}\right)\frac{1}{\alpha n_s^{-1/3}}\frac{1}{g_a\bar{t}} = 0.0579\frac{1}{\alpha n_s^{-1/3}}\frac{1}{g_a\bar{t}} \quad (16)$$

Direct computation of $g_a\bar{t}$ via eq 14 is straightforward and will be performed below. However, it is useful to approximately evaluate $g_a\bar{t}$ as follows. The integrands in eq 14 are dominated by the double exponential functions, which exhibit nearly step-function behavior

$$\begin{aligned} \mathrm{e}^{-\mathrm{e}^{\tilde{x}-\tilde{r}}} &\approx 0 \quad \tilde{x} > \tilde{r} \\ \mathrm{e}^{-\mathrm{e}^{\tilde{x}-\tilde{r}}} &\approx 1 \quad \tilde{x} < \tilde{r} \end{aligned} \quad (17)$$

Thus,

$$\begin{aligned} \int_{\tilde{r}}^\infty \mathrm{d}\tilde{r}'\tilde{r}'^2(1-\mathrm{e}^{-\mathrm{e}^{\tilde{x}-\tilde{r}'}}) &\approx \frac{1}{3}(\tilde{x}^3 - \tilde{r}^3) \quad \tilde{x} > \tilde{r} \\ &\approx 0 \qquad\qquad \tilde{x} < \tilde{r} \end{aligned} \quad (18)$$

Using eqs 17 and 18 in eq 14 and changing the order of integrations over $\tilde{x}$ and $\tilde{r}$ leads to the very simple result

$$g_a\bar{t} = \eta \int_0^{\tilde{r}_c} \mathrm{d}\tilde{r}\tilde{r}^2\, \mathrm{e}^{\tilde{r}-\eta\tilde{r}^3/3} \quad (19)$$

A similar result without the cutoff was derived by Scher and Lax.[1] Without the cutoff, the integral can be evaluated by the saddle-point method and the asymptotic dependence of $g_a\bar{t}$ on η is found to be[1]

$$g_a\bar{t} \approx \eta^{-1/4}\, \mathrm{e}^{2\eta^{-1/2}/3} \quad (20)$$

with the conductivity also exponential in $\eta^{-1/2}$. The dependence of the conductivity on η within the percolation analysis is $\mathrm{e}^{-A\eta^{-1/3}}$, and hence the CTRW result following from eq 20 predicts the incorrect scaling with site density and gives rise to substantial differences in the magnitude of the DC conductivity with decreasing site density.

4. Results and Comparison to a Percolation-Based Model

An analytic model for the DC conductivity was derived by Butcher et al.[3,4] following the Miller and Abrahams model,[11] in which the conductivity is equivalent to that of a related conductance network, and using the percolation analogies drawn by Ambegoakar, Halperin, and Langer,[9] and others. The dominant contribution to the conductivity arises from those critical conductances that allow for a macroscopic connected path of conductances below the critical value. Simulation data indicate that the percolation radius r_p, required such that a macroscopic conduction path of sites exists along which all site separations are less than or equal to r_p, is given by

$$\begin{aligned} r_p &= \left(\frac{3N_p}{4\pi n_s}\right)^{1/3} \\ \tilde{r}_p &= (3N_p)^{1/3}\, \eta^{-1/3} \end{aligned} \quad (21)$$

where N_p is the average number of neighboring sites to which a typical site is connected at percolation. In 3d, simulations indicate that $N_p \approx 2.7$ and hence $\tilde{r}_p = 2.008\eta^{-1/3}$. The normalized DC conductivity is predicted to be[3,4]

$$\tilde{\sigma}_{\mathrm{DC}} = \sigma_p \mathrm{e}^{-2\alpha r_p} \quad (22)$$

with the coefficient σ_p given by

$$\begin{aligned} \sigma_p &= \frac{2\pi}{3}\frac{n_s^2}{\alpha}\left[\int_0^{r_p} \mathrm{d}r r^2 r_p^2 \mathrm{e}^{-2\alpha|r-r_p|} + \int_{r_p}^\infty \mathrm{d}r r^4 \mathrm{e}^{-2\alpha|r-r_p|}\right] \\ &= \frac{\pi}{6}\left(\frac{3N_p}{4\pi R_p^3}\right)^2 (3 + 6R_p + 7R_p^3 + 4R_p^4 - R_p^2 \mathrm{e}^{-2R_p}) \end{aligned} \quad (23)$$

where $R_p = \alpha r_p$. The predictions of eqs 22 and 23 are in very good agreement with simulation data, and will be used below to approximate the "exact" DC conductivity. For low site densities $R_p \gg 1$, the conductivity becomes

$$\tilde{\sigma}_{DC} = \frac{3^{1/3}N_p^{4/3}}{2\pi}\eta^{2/3}\, e^{-(3Np)^{1/3}\eta^{-1/3}} \quad (24)$$

The above percolation analysis suggests a transition rate cutoff of $\tilde{r}_p$, or more generally a cutoff radius scaling as $\tilde{r}_c = (3N_p)^{1/3}\eta^{-1/3}$ where N_p can be considered adjustable. An analytic asymptotic low-density expression for the DC conductivity within the modified CTRW model can be obtained through evaluation of eq 19. At low densities, $\tilde{r}_c \gg \eta\tilde{r}_c^3/3$, and hence the latter term in the exponential of eq 19, can be treated as a constant. Then,

$$g_a\bar{t} \approx \eta \int_0^{\tilde{r}_c} d\tilde{r}\tilde{r}^2\, e^{\tilde{r}-(1/3)\eta\tilde{r}_c^3} = \eta e^{-1/3\eta\tilde{r}_c^3}[(\tilde{r}_c^{\,2} - 2\tilde{r}_c + 2)\, e^{\tilde{r}_c} - 2] \quad (25)$$

Neglecting the last term of -2 in eq 25, the DC conductivity is, following from eq 16 and some algebra,

$$\sigma_{DC} \approx 0.0498[(3N_p)^{2/3} - 2(3N_p)^{1/3}\eta^{1/3} + 2\eta^{2/3}]^{-1}\, e^{N_p}\, e^{-(3N_p)^{1/3}\eta^{-1/3}}$$

$$\approx \frac{0.05}{(3N_p)^{2/3}}\, e^{N_p}\, e^{-(3N_p)^{1/3}\eta^{-1/3}} \quad (26)$$

This form is quite accurate over a wide range of densities. Using the cutoff radius $\tilde{r}_c = \tilde{r}_p$, or equivalently $N_p = 2.7$ as suggested by the percolation-based models, renders the asymptotic exponents in the modified CTRW result of eq 26 and the percolation result of eq 24 identical. However, the numerical prefactors are rather different, with the modified CTRW approach having only a vanishing density dependence in the prefactor.

The normalized conductivity predicted by eq 16 via numerical integration of eq 14, and by eqs 22 and 23, is shown in Figure 2 on a log scale as a function of the normalized inverse site density parameter $\alpha n_s^{-1/3}$ ($\eta = \pi/2(\alpha n_s^{-1/3})^{-3}$). Also shown is the usual CTRW result, for which there is no cutoff. The usual CTRW model exhibits the anticipated orders-of-magnitude deviations at low densities. The modified CTRW naturally predicts a larger conductivity than the CTRW at lower site densities $\alpha n_s^{-1/3}$ and predicts a DC conductivity roughly 1 order of magnitude larger than that found in the simulation/percolation results. The difference in prefactors between eqs 24 and 26 also shows that the deviation grows slowly with decreasing density. At high site densities, the existence of a cutoff in the transition spectrum is not important, and the CTRW and modified CTRW values become quite similar and are typically slightly lower than the "exact" values. Thus, the modified CTRW rectifies the low-density problem of the usual CTRW but does not affect the high-density regime.

The modified CTRW does not treat the DC conductivity as a percolation problem, and hence the appropriate "cutoff" transition rate is not necessarily $\tilde{r}_c = \tilde{r}_p$ or $N_p = 2.7$. Increasing $\tilde{r}_c$ to $\tilde{r}_c = 2.3\eta^{-1/3}$ or $N_p = 4.05$ has been found to yield excellent results. The modified CTRW prediction of the DC conductivity for $\tilde{r}_c = 2.3\eta^{-1/3}$ is shown in Figure 3 vs the normalized inverse site density parameter $\alpha n_s^{-1/3}$, along with the analytic result of eqs 22 and 23. The agreement between the two is extremely good at lower densities down to $\alpha n_s^{-1/3} = 24$ or $\eta = 0.00011$. Furthermore, with this cutoff value less than 2% of the sites are eliminated from consideration due to being "too isolated" from their neighbors. The modified CTRW with this cutoff represents a minor adjustment to the topology of the problem,

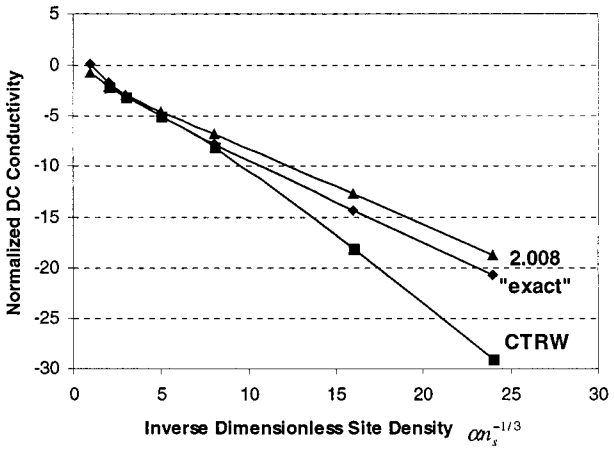

Figure 2. Normalized DC conductivity $\tilde{\sigma}_{DC}$ versus inverse site density, as predicted by the CTRW, the modified CTRW with $r_c = 2.008\eta^{-1/3}$, and by the nearly exact analytic expression of Butcher et al.

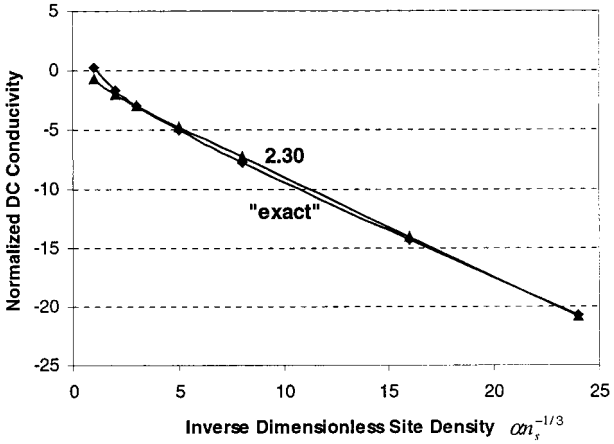

Figure 3. Normalized DC conductivity versus inverse site density, as predicted by the modified CTRW with $r_c = 2.3\eta^{-1/3}$, and by the nearly exact analytic expression of Butcher et al.[3,4]

by neglecting a very small fraction of "special" isolated sites, and yet rectifies the one major limitation of the original CTRW approach.

5. Discussion

Connectivity is a major factor in determining conductivity at low site densities. The percolation approach considers only that connectivity necessary to obtain the one percolation path, such that the conductivity is dominated by the largest impedance in that one path. At low densities, it is clear that such an approach is relevant. The CTRW retains the full connectivity of a regular lattice and thus incorporates the connectivity necessary to predict finite-frequency/finite-time behavior, with the disorder subjugated into an effective transition rate. To obtain the transition rate, the usual CTRW essentially averages the transition rates equally. Such an averaging thus builds in a broad spectrum of rates that forces, probabilistically, the sampling of paths containing the extremely slow transition rates. The modification introduced here of eliminating a very few of the slowest transition rates can thus be viewed as a biased sampling of the available rates in which hopping transitions that will statistically never occur even in the DC percolation problem are precluded. It is rather remarkable that such a simple modification to the CTRW method, incorporating only the one key feature of the geometry of the percolation model, can lead to very good predictions for the DC conductivity.

The ad-hoc adjustment of the cutoff radius from $\tilde{r}_p$ to $1.15\tilde{r}_p$ was proposed with no sound physical basis. One rationale for this small increase in the cutoff is that even within the percolation problem one must go slightly beyond the percolation point to obtain finite DC conductivity. As the percolation radius is increased slightly, a vastly larger number of possible paths become reasonable parallel conduction paths and the conductivity increases from zero to finite values. In the analytic model of Butcher et al., impedances with separation larger than $\tilde{r}_p$ are also permitted (see eq 23), but within only a narrow range scaling with the factor α. The analytic result of eq 26 for the modified CTRW shows, however, that the exponent in the conductivity is clearly increased relative to the percolation value, and hence the modified CTRW result with the increased cutoff radius cannot give the exact asymptotic conductivity behavior. Nonetheless, it is of considerable interest that such strong cancellations between a modified exponent and a modified prefactor can occur to yield very accurate numerical results over essentially the entire range of physically accessible densities.

The AC conductivity depends on hopping across finite distances among sites in the disordered system. The long-range percolation path that ultimately controls the DC conductivity thus plays a negligible role in the AC conductivity at frequencies above that for which the original CTRW AC conductivity is larger than the percolation DC conductivity. The introduction of a cutoff in the near-neighbor transition rate alters the AC conductivity only in the low-frequency range where it falls below the percolation DC conductivity. At the higher frequencies, the limitation in transitions becomes unimportant. Hence, the AC conductivity predicted by the modified CTRW model here is expected to retain the excellent predictions of the original CTRW at moderate to high frequencies.[2]

At high densities, the CTRW predictions have been found to agree well with experimental data even in the DC limit.[1] However, there are some deviations between the CTRW and the simulated DC conductivity value at high densities, with the simulation/percolation-based models predicting higher conductivity (see Figure 1). For computational tractability, the simulation studies consider conductances between sites only within some finite range. The number of "parallel" channels of conduction is thus restricted, and this could be an issue at the highest site densities. However, the simulation predictions yield a larger DC conductivity than predicted by the CTRW, which is counter to the trend expected from the above limitation of the simulation results. Therefore, the modest deviations between the CTRW and simulation/percolation models at high densities remain unsolved.

In summary, a modified CTRW approach has been proposed in which the near-neighbor transition rate spectrum is truncated at a critical distance r_c. The resulting predictions for DC conductivity agree very well with simulation data when the critical distance is chosen to be only slightly larger than the critical percolation distance. With the retention of the excellent high-frequency behavior of the original CTRW, the modified approach bridges the gap between DC and AC conductivities and thus provides a single framework within which the conductivity of disordered systems can be accurately accessed.

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