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LETTER TO THE EDITOR

Numerical calculations of non-Ohmic hopping conductivity in one-dimensional systems

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Abstract. The current–field characteristic of a 1D, variable-range hopping system is calculated for the first time by solving non-linear electron conservation equations. A previous linearised calculation is shown to violate total electron number conservation. This condition is imposed in the non-linear calculation together with appropriate modifications of the contact boundary conditions. For weak fields these changes have no effect on the results but for large fields there are large changes in the calculated values of the current. The standard deviation of the fluctuations of $\log J$ produced by varying the chemical potential μ is shown to decrease with increasing field. Previous calculations of the variation of $\langle \log J \rangle_\mu$ with the number of sites, N, are extended to $N = 10^4$. Fluctuations of $\langle \log J \rangle_\mu$ with increasing N are found, ~ 0.5, about a systematic decrease towards what appears to be an asymptotic limit.

This investigation was prompted by the non-Ohmic current–voltage characteristics observed in 1D hopping systems by Webb [1, 2] and calculated for 3D hopping systems by Böttger and co-workers [3–5]. The calculations in reference [3] are for 'R-hopping', i.e. the electron energies are assumed to be the same on all the hopping sites. References [4] and [5] are for the more realistic case of 'R–ε-hopping' in which the electron energies vary from site to site. However the calculations in [4] and [5] are made for numbers of sites $N \leqslant 500$ which is very small for a 3D model and, to further simplify the calculations, the hopping rate is given the form appropriate to strong electron–phonon coupling. In the more usual case of weak electron–phonon coupling a more complicated rate is required. In this paper we outline some preliminary numerical results for 1D hopping systems for the case of weak electron–phonon coupling. The restriction to 1D makes it easier to simulate macroscopic samples with $N = 10^3$–10^4.

In a previous paper [6] two of the authors investigated Ohmic behaviour for such a model using a Miller–Abrahams conductance network [7] in which the voltage is set equal to $V_{L(R)}$ at all the sites within 5% of the length of the sample at the left-hand (right-hand) end. These are contact regions and the voltages assumed in them make the occupation probability f_m of the mth contact site independent of the voltage drop across the sample. In the non-linear regime we also began by assuming that f_m in the contacts is independent of applied field. However, this procedure gives results which do not conserve the overall number of electrons in the sample. To achieve such conservation

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we employ here boundary conditions of the type originally introduced by Böttger and Wegener [3]. However, to simplify their application, we suppose that all the contact sites have the same energy. Then the contact boundary condition reduces to $f_m = f$, independent of m, for all sites m in both contacts. For 'interior' sites m lying between the contact regions the energy ε_m is taken to be a random variable lying in the range $(-W/2, W/2)$.

The question arises as to whether the new boundary conditions yield the same results in the Ohmic regime as the old ones. The answer would be expected to be in the affirmative, because, although the old results violate overall electron number conservation, they do so only by an infinitesimal amount in the Ohmic regime which should not affect the conductivity obtained by solving the non-linear equations with the new boundary conditions for low fields. We also extend the old calculations of the size dependence of the Ohmic conductivity up to $N = 10^4$. Finally in the non-Ohmic regime, we calculate the field dependence of the fluctuations of the current with varying chemical potential.

We consider a system of hopping sites randomly located on a straight line at the points x_m where m is an integer labelling the sites. An electron on site m has an energy which we denote by ε_m. The values of ε_m are selected as described above. The hopping rate between sites m and n at temperature T is given by

$$R_{mn} = R_0 \exp(y_{mn} - 2\alpha|x_{mn}|)/2\sinh|y_{mn}| \tag{1}$$

where

$$x_{mn} = x_m - x_n \tag{2}$$

$$y_{mn} = y_m - y_n \tag{3}$$

$$y_m = (\varepsilon_m + eEx_m)/2k_B T. \tag{4}$$

Here R_0 is only weakly temperature-dependent in comparison with the exponential factors in (1) and in the numerical calculations, R_0 is set equal to a constant. The quantity α^{-1} is the localization length, k_B is Boltzmann's constant, E is an applied electric field and the electrical charge $e = |e|$.

The particle current from site m to site n is given by

$$\begin{aligned} j_{mn} &= f_m(1-f_n)R_{mn} - f_n(1-f_m)R_{nm} \\ &= W_{mn}[f_m(1-f_n)\exp(y_{mn}) - f_n(1-f_m)\exp(y_{nm})] \end{aligned} \tag{5}$$

where

$$W_{mn} = R_0 \exp(-2\alpha|x_{mn}|)/2\sinh|y_{mn}| = W_{nm} \tag{6}$$

We consider a steady state and at each 'interior' site m (i.e. when x_m is in the part of the line between the contacts) we have particle conservation:

$$\sum_n j_{mn} = 0. \tag{7}$$

Moreover, the total particle current is

$$J = \sum_{m<m^*} \sum_{n\geq m^*} j_{mn} \tag{8}$$

where m^* is any fixed interior site.

The quantities f_m for all interior sites may be determined from equation (7) if all the f_m values are known in the exterior (contact) sites. We have taken all sites within 10% of each end of the system to constitute 'contact regions' (CR). The values of f_m in these regions are denoted f_L and f_R for the left- and right-hand contacts respectively. Following Böttger and Wegener we set $f_L = f_R$. We then need $I + 1$ equations to solve for the f_m values where I is the number of interior sites. The I interior equations are those included in equation (7), and the $(I + 1)$th equation expresses the conservation of overall electron number

$$\sum_m f_m = \sum_m f_m^0 \tag{9}$$

with

$$f_m^0 = [\tfrac{1}{2}\exp\{-(\varepsilon_m - \mu)/k_B T\} + 1]^{-1} \tag{10}$$

denoting the equilibrium value of the occupation probability of site m when the Fermi level is μ. The summations of equation (9) are over the sites in the contacts as well as in the interior.

The numerical problem to be solved is reduced to a set of non-linear equations

$$F_i(f_1^{k+1}, \ldots, f_{i-1}^{k+1}, f_i \ldots, f_N^k) = 0 \qquad i = 1 \ldots N \tag{11}$$

in which k identifies the iteration number. For any particular iteration equations (11) may be solved for f_i, $i = 1, \ldots N$ and f_i^{k+1} set equal to f_i. This procedure forms the basic step of a non-linear Gauss–Seidel iteration which may be generalised to a non-linear successive over-relaxation (SOR) method in which we set $f_i^{k+1} = f_i^k + \omega_k(f_i - f_i^k)$. Here ω_k is a suitable relaxation parameter. If we restrict ourselves to a one-step SOR method the explicit form becomes

$$f_i^{k+1} = f_i^k - \omega D^{-1} F_i(f^{k,i}) \qquad i = 1, \ldots N \qquad k = 0, 1, \ldots \tag{12}$$

where

$$f^{k,i} = [f_i^{k+1}, \ldots f_{i-1}^{k+1}, f_i^k, \ldots f_N^k]^T \tag{13a}$$

with T denoting the transpose of the vector $f^{k,i}$ and k the iteration number. In equation (12)

$$D = \partial_i F_i(f^{k,i}) \tag{13b}$$

which is explicitly given by

$$D = [F_i(f^{k,i}) - F\{f^{k,i} - F_i(f^{k,i})u^i\}]/F_i(f^{k,i}) \tag{13c}$$

where u^i is a vector in which the ith element is set equal to 1 and all other elements are set equal to 0. The set of equations defined by equation (12) have been solved on a CRAY X-MP/48 at the Rutherford–Appleton Laboratory. The computations proceed at a sustainable rate of 141 Mflops.

Throughout the calculations the energy values are chosen randomly in the range $-W/2$ to $+W/2$ with $W = 1$ meV, the localization length $\alpha^{-1} = 10^2$ nm and the average intersite spacing $a = 2$ nm. Then the sample length is $L = aN$, where N is the number of sites. To present the results we introduce reduced variables

$$T^* = k_B T/W \tag{14a}$$

$$E^* = eEL/W \tag{14b}$$

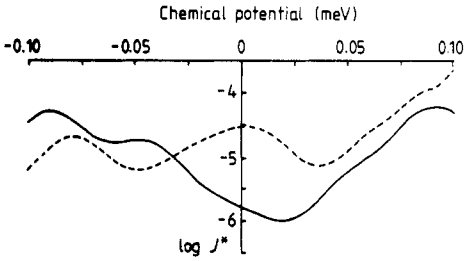

Figure 1. $\log J^*$ against μ for $E^* = 10^{-5}$, $T^* = 0.008$ and $N = 1000$; ———: particles are conserved and CR = 10%; — — —: particles are not conserved and CR = 10% coincides exactly with full curve; – – –: particles are conserved and CR = 5%.

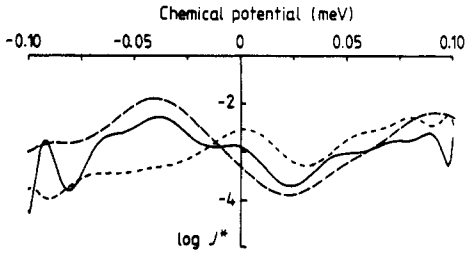

Figure 2. $\log J^*$ against μ for $E^* = 10^{-3}$, $T^* = 0.008$ and $N = 1000$; ———: particles are conserved and CR = 10%; — — —: particles are not conserved and CR = 10%; – – –: particles are conserved and CR = 5%.

$$J^* = J/R_0 \tag{14c}$$

and set $T^* = 0.008$ which is the largest value used in reference [6]. The energy scale of the current fluctuations is determined by $k_B T$. The use of a relatively high temperature means that we may carry out the computation with a reasonably coarse interval for μ. Steps of $k_B T/2$ are fine enough to prevent both loss of structure and the imposition of artificial structure.

In figure 1 we plot $\log J^*$ against μ for $E^* = 10^{-5}$ with $N = 10^3$. The full line is obtained using equation (9) for CR = 10%. It conceals another plot obtained using the old boundary conditions for CR = 10%. With $E^* = 10^{-5}$ we are well down in the Ohmic region of the current–field characteristic. We see that, in this region, the calculated values of J^* are insensitive to the imposition of total electron number conservation. They are actually much more sensitive to the extent of the contact regions which affects the hopping rates taken into account in the interior of the line. This is illustrated by the dotted line which is drawn for CR = 5% when the new boundary conditions are imposed.

Figure 2 shows similar results when $E^* = 10^{-3}$. The full line is for CR = 10% and the dotted line is for CR = 5%, both obtained using equation (9). The difference between these two plots is similar to that found in figure 1 when $E^* = 10^{-5}$. The dashed curve in figure 2 is obtained with CR = 10% by replacing equation (9) by the old boundary condition $f_m = f_m^0$ in the contacts. We see that it differs significantly from the full line. With $E^* = 10^{-3}$ we have moved out of the Ohmic regime and the imposition of total electron number conservation makes a significant difference to the results.

In figure 3 we give similar results for $E^* = 10$, 10^{-2} and 10^{-5}. Total electron number conservation is imposed in all cases. The figure shows the field dependence of the fluctuations as the field E^* moves out of the Ohmic region ($E^* = 10^{-5}$) through intermediate values ($E^* = 10^{-2}$) to large value ($E^* = 10$). We see that the fluctuations smooth out as E^* increases. This observation is confirmed in figure 4 which is a plot of the standard deviation of the fluctuations of $\log J^*$ as a function of $\log E^*$. The standard deviation itself shows fluctuations (full curve) but when these are smoothed out (broken curve) we find a significant decrease from 0.6 when $E^* = 10^{-5}$ to <0.1 when $E^* > 1$. Finally, in figure 5 we plot $\langle \log J^* \rangle_\mu$ against the number of sites N. Here, the angle brackets indicate an average over μ (51 values). Again there are significant fluctuations (full curve) but a definite trend towards what appears to be an asymptotic limit is found when the fluctuations are smoothed out (broken curve).

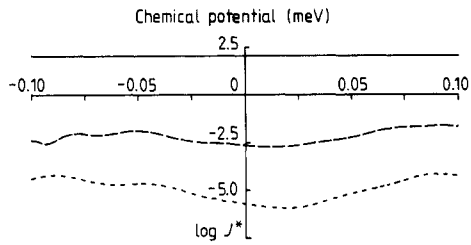

Figure 3. $\log J^*$ against μ for $T^* = 0.008$, $N = 1000$, CR = 10% with particles conserved when $E^* = 10$ (———); $E^* = 10^{-2}$ (— — —) and $E^* = 10^{-5}$ (– – –).

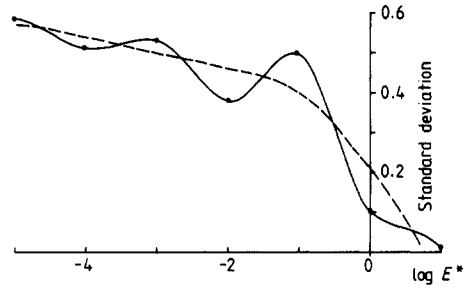

Figure 4. A plot against $\log E^*$ of the standard deviation of the fluctuations of $\log J^*$ obtained by varying the chemical potential, (CR = 10%, $T^* = 0.008$).

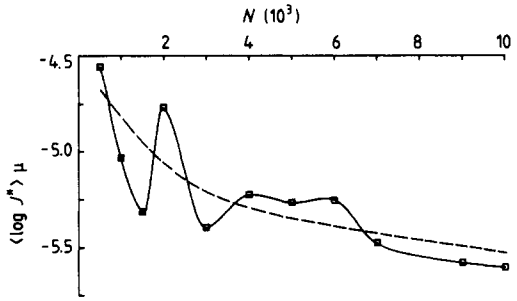

Figure 5. $\langle \log J^* \rangle_\mu$ against the number of sites N for $T^* = 0.008$.

These preliminary results are easily extended to discuss the behaviour of the current–field characteristic in the non-Ohmic regime. We discuss that in another paper. The purpose of the present communication is threefold. Firstly, to show that the previous neglect of total electron number conservation in the Ohmic regime did not affect the published results. Secondly, to show that this conservation law is important in the non-Ohmic regime and thirdly, to show that the extent of current fluctuations with varying Fermi level becomes smaller at high fields.

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References

[1] Webb R A, Harstein A, Wainer J J and Fowler A B 1985 *Phys. Rev. Lett.* **54** 1577
[2] Webb R A, Fowler A B, Harstein A and Wainer J J 1986 *Surf. Sci.* **170** 14
[3] Böttger H and Wegener D 1984 *Phil. Mag.* **50** 409
[4] Böttger H, Szyler R and Wegener D 1985 *Phys. Status Solidi* b **128** 4179
[5] Böttger H, Szyler R and Wegener D 1986 *Phys. Status Solidi* b **133** 4143
[6] McInnes J A and Butcher P N 1985 *J. Phys. C: Solid State Phys.* **18** L921
[7] Miller A and Abrahams S 1960 *Phys. Rev.* **120** 745