Non-Ohmic Variable-Range Hopping Transport in One-Dimensional Conductors

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We investigate theoretically the effect of a finite electric field on the resistivity of a disordered one-dimensional system in the variable-range hopping regime. We find that at low fields the transport is inhibited by rare fluctuations in the random distribution of localized states that create high-resistance breaks in the hopping network. As the field increases, the breaks become less resistive. In strong fields the breaks are overrun and the electron distribution function is driven far from equilibrium. The logarithm of the resistance initially shows a simple exponential drop with the field, followed by a logarithmic dependence, and finally, by an inverse square-root law.

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Motivation and main results.—Potential applications in nanotechnology have generated a widespread interest in one-dimensional (1D) conductors, including carbon nanotubes, nanowires, and conducting molecules. Before such applications may become practical, a number of fundamental physics questions need to be resolved. One of them is the mechanism of the non-Ohmic hopping transport, observed in a number of recent experiments [3–5]. Especially interesting and challenging is the ultimate low-T limit where variable-range hopping (VRH), i.e., hopping between distant localized states dominates. Remarkably, this problem has long remained untouched by theorists except for one early numerical study [6]. Recently, however, the following analytical formula has been proposed [7]:

$$\rho \propto \exp(Ck_BT_0/\gamma a)^{1/2},$$

where $\rho$ is the resistivity, $F = e\mathcal{E}$ is the product of the electron’s charge and the electric field, $a$ is the localization length, $T_0$ a parameter of dimension of temperature defined below, and $C$ is a numerical coefficient left undetermined. This formula has immediately become a matter of controversy because in its derivation a very important and non-trivial peculiarity of the 1D geometry was overlooked: unavoidable highly resistive segments (“breaks”) on the conducting path [8–10]. In an attempt to include this physics, Eq. (1) was subsequently revised in a follow-up article [11]. Yet the actual situation is more intriguing. As demonstrated below, the problem has quite a rich regime diagram. The resistivity may take three different functional forms, depending on the strength of the electric field (weak, intermediate, or strong). It is Eq. (1) that proves to be correct in strong fields, $F \gg k_BT/a$, not its subsequent revision [11]. We compute the exact coefficient in Eq. (1) to be $C = 8$. The main message of our analysis is that the role of the breaks diminishes and eventually becomes insignificant as the electric field increases. We hope that these ideas and new results may provide a much needed theoretical basis for the purposeful experimental study of the non-Ohmic VRH expected to be reported in the near future.

Model.—Since the problem has proved to be rather delicate, in this study we approach it within a simplified model of noninteracting electrons. (In real systems interaction effects can be suppressed by placing the 1D conductor near a metallic gate.) The localized states are regarded as random points $(x_j, \epsilon_j)$ with the Poisson distribution of a constant density $g$ in the position-energy space. This is justified at low $T$ where electron transport is determined by a narrow range of energies. Similarly, at low enough $T$ we can neglect the field dependence of the localization length $a$ because the electric fields of interest, say, $F \sim k_BT/a$, are small.

The rate of hopping $\Gamma_{jk}$ from site $j$ to site $k$ is given by the usual model form [12]

$$\frac{1}{\gamma} = f_j(1 - f_k) \exp\left[\frac{-2}{a} |x_j - x_k| - \frac{\max(D_{jk}, 0)}{k_BT} \right],$$

where $\gamma$ of dimension of frequency is determined by the electron-phonon coupling, $0 < f_j < 1$ is the occupation factor of the $j$th site, and $D_{jk} = \epsilon_k - \epsilon_j - F(x_k - x_j)$. The net current from $j$ to $k$ is given by $I_{kj} = e(\Gamma_{jk} - \Gamma_{kj})$. The transport problem amounts to solving the current conservation equations $\sum_{k \neq j} I_{kj} = 0$ for $f_j$’s at a given fixed $F$.

One conventional way to present the result is in the form

$$f_j = \left[\exp(\epsilon_j - \mu_j/k_BT) + 1\right]^{-1},$$

where $\mu_j$ is termed the chemical potential of the $j$th site. In the most relevant situation, $\Delta_{jk} = |\epsilon_j - \epsilon_k| + |\epsilon_j - \mu_j| + |\epsilon_j - \epsilon_k| \gg k_BT$, the currents $I_{kj}$ are given by [13]

$$I_{kj} \approx e \gamma \exp\left(\frac{-2}{a} |x_k - x_j| - \frac{\Delta_{jk}}{2k_BT} \right) \sinh \frac{\epsilon_j - \epsilon_k}{k_BT},$$
Thus, the logarithm of the resistance of a typical link is

\[ \ln R_{ij} = \xi_j - \xi_i \]

in concentration.

The resistances \( R_{ij} \) are locally absent in a large area where the angle brackets denote averaging over a set of questions: What is the highest \( F \) when Eq. (5) still applies? Do breaks continue to play a role at larger \( F \)? How does \( \rho \) depend on \( F \) at such fields? Our answers to these questions are given below.

Intermediate fields.—We begin by showing that the region of validity of Eq. (5) is very limited. Indeed, in the derivation it was assumed that the voltage drop across each link is less than \( k_B T/e \). However the voltage drop on every optimal break is equal to \( IR_{\Omega} = (F/e \rho)R_{\Omega} = F/eN_{\Omega} \); therefore, Eq. (5) is justified only at \( F \ll F_{\Omega} = k_B T/N_{\Omega} \). To compute \( \rho \) at larger \( F \) a different approach is needed. Our strategy resembles that of Ref. [12]. We assume that a given fixed current \( I \) flows through the system and compute \( F = (d\xi/dx) \). Then we find \( \rho \) from the relation

\[ \ln \left[ \frac{\rho(F, T)}{(R_0/a)} \right] = u_I - \ln \left( \frac{k_B T}{F a} \right) \cdot u_I = \ln \left( \frac{e \mu}{T} \right). \]  

At this stage we focus on the intermediate field (current) regime, defined by \( u_M \ll u_I \ll u_M \). Let \( \Delta \xi_i \equiv \xi_i - \xi_{i+1} \) be the electrochemical potential difference across the \( i \)th link of the optimal path. Since \( x_M \) is the correct estimate of the typical length of the link, we have \( F \sim (\Delta \xi_i)/x_M \), where the angle brackets denote averaging over \( i \). Introducing \( P_\xi \), the probability distribution function (PDF) of the random quantity \( \Delta \xi_i \), we can write this as

\[ F \sim \frac{1}{x_M} \int_0^\infty P_\xi(\Delta \xi)\Delta \xi d\Delta \xi. \]  

Let us establish the functional form of \( P_\xi \). The behavior of \( P_\xi(\Delta \xi) \) at \( \Delta \xi \ll k_B T \) is determined by links with Ohmic resistances \( R_\Omega e^u \), where \( u \ll u_I \). They remain operating in the Ohmic regime, and so for them \( \Delta \xi_i/k_B T = \exp(u - u_I) \ll 1 \). Using RR’s results for the PDF of the (diamond-shaped) Ohmic breaks, we obtain

\[ P_\xi = \frac{C}{\Delta \xi} \exp \left( -\frac{T}{2T_0} \left[ u_I - \ln \left( \frac{k_B T}{\Delta \xi} \right) \right]^2 \right). \]  

where \( 1 \ll (k_B T/\Delta \xi) < u_I - u_M \) and \( C \) is a prefactor with a subleading dependence (presumably, a logarithmic one) on \( \Delta \xi \) and \( T \). Note that the typical links, \( u \ll u_M \), are included in Eq. (8) [they correspond to the lowest \( \Delta \xi \ll k_B T \exp(u_I - u_I) \) at which this formula is still valid]. Consider now the links with \( \Delta \xi_i \gg k_B T \), which are necessarily atypical, i.e., some sort of breaks. The geometry of such non-Ohmic breaks can be established as follows. In a vicinity of a non-Ohmic break the largest voltage variation is across this break; therefore, the electrochemical potential has nearly the same value, say, \( \xi_i \), on the sites adjacent to the left boundary of the break and another value, \( \xi_{i+1} \) for those on the right side. Assuming that \( \Delta \xi_i = \Delta \mu_j \) (borne

\[ \xi_j = \mu_j - Fx_j \]

is the electrochemical potential. Knowing \( \mu_j \)'s (or \( f_j \)'s), one then computes the total current \( I \) and the resistivity \( \rho(F, T) = eI/F \).

Ohmic regime.—In general, the above transport equations are nonlinear in \( \mu_j \) but in the Ohmic limit, \( F \to 0 \), \( T > 0 \), both \( \mu_j \) and \( \xi_j - \xi_i \) are smaller than \( T \) and Eq. (3) becomes \( (\xi_j - \xi_i)/e = I_{jk}R_{jk} \), where \( R_{jk} = R_0 \times \exp(2/\alpha)|x_j - x_i + (\Delta_j/k_B T)| \). Therefore, the system is equivalent to the network of resistors \( R_{jk} \) while \( \xi_j/e \) is simply the voltage at the \( j \) node [1,2]. Here \( R_0 = k_B T/(e^2 \gamma) \) and \( \Delta_j \) is to be computed at \( \mu_j = \mu_k = 0 \). The resistances \( R_{jk} \) differ by many orders of magnitude from one another and this is what makes the problem nontrivial. Let us review the basic ideas developed for its solution. A crucial step [1,2] is to realize that among many possible current paths through the network, a single optimal one heavily dominates the conduction. In one dimension this path has the topology of a linear chain, so that we can label the nodes of the path consecutively: \( 1, 2, \ldots, i, i + 1 \), etc. The typical separation in position \( (x_M) \) and in energy \( (\epsilon_M) \) between these nodes follows from the conventional Mott’s argument [1,2]:

\[ \frac{\epsilon_M(T)}{k_B T} = \frac{x_M(T)}{a} = u_M = \sqrt{\frac{T_0}{T}} k_B T_0 = \frac{1}{ga}. \]  

Thus, the logarithm of the resistance of a typical link is

\[ \ln R_{i,i+1} \sim x_M/a + \epsilon_M/k_B T - u_M. \]  

In addition to typical links, there are places in the conductor where hopping sites are locally absent in a large area \( A \) of the \((x, \epsilon)\) plane. The probability of such breaks is very small, \( \exp(-gA) \), but its resistance far exceeds that of a typical link. A careful analysis of Raikh and Ruzin (RR) [10] showed that the dominant contribution to \( \rho \) is provided by breaks that are shaped as diamonds and have the sizes \( u_0 a/2 \) and \( 2T_0 \) in the position and energy dimensions, respectively, see Fig. 1(a) [14]. Here \( u_0 = T_0/T \). These “optimal” breaks have resistances \( R_0 = R_0 \exp(u_0) \) and are present in concentration \( N_0 \sim \exp(-u_0/2)/x_M \), which yields \( \rho \sim R_0 N_0 \), i.e., [10]

\[ \ln \rho(0, T) \approx T_0/2T, \quad T \ll T_0. \]  

As customary in this field, we will not attempt to compute the preexponential factor of the resistivity [15].

FIG. 1. (a) Ohmic and (b) non-Ohmic breaks. The dots represent hopping sites. The horizontal thin lines are the links of the optimal path carrying the current across the breaks. The width of each break is \( a u_I/2 \).
out by the results) we see from Eq. (3) that the current through a pair of sites $j$ and $k$ located on the opposite sides of the break is $I_{jk} \approx e\gamma \exp(- (2/\alpha)|x_j - x_k| - (\Delta E_j/2kT) + (\Delta E_k/2kT))$. The shape of the non-Ohmic break should be such that the inequality $I_{jk} \ll 1$ is obeyed.

The optimal, i.e., the most probable breaks are those of minimal area. Using only elementary geometrical considerations one readily finds that the optimal shapes comprise a continuum of curvilinear hexagons of area $A_k = au_k (k_BT u_k + \Delta E_k)/2$, see Fig. 1(b). Hence, $P_k(\Delta E_k) \sim \exp(-gA_k)$ for $\Delta E_k \gg k_BT$, or, more accurately,

$$P_k = \frac{\mathcal{C}}{k_BT} \exp\left(-\frac{T u_k^2}{2T_0} - \frac{u_k \Delta E_k}{2k_BT_0}\right), \quad \Delta E_k \gg k_BT. \quad (9)$$

Eqs. (8) and (9) match by the order of magnitude at $\Delta E_k \sim k_BT$. Substituting them into Eq. (7), we find the main voltage generators in the system are the non-Ohmic breaks with $\Delta E_k \sim k_BT u_k$. For $F$ we obtain

$$F \sim (Ck_BT/au_k^2) \exp(-u_k^2/2u_M^2). \quad (10)$$

Solving this for $u_I$ and using Eq. (6), we arrive at

$$\ln \rho = \sqrt{\frac{2T_0}{T}} \ln\left(\frac{\nu k_BT}{F_a}\right) - \ln k_BT/a, \quad F_\Omega \ll F \ll \frac{k_BT}{a}, \quad (11)$$

where $\nu = Cu_k^2/u_M^2$. The main message of this analysis is that atypically large resistors are progressively eliminated as $F$ increases ($u_I$ decreases). Note that at $u_I \sim u_M$ the distinction between breaks and typical links disappears, so that the result $\Delta E_k \sim k_BT u_k$ for the breaks is a good estimate of the overall $\langle \Delta E \rangle$. Thus, at this point $F \sim k_BT/a$ and therefore $\nu \sim C \sim 1$.

**Strong fields.**—A quick if unilluminating derivation of $\rho$ at $F \gg k_BT/a$ assumes that Eq. (10) holds throughout the non-Ohmic regime. In strong fields, where $u_I \ll u_M$, the exponential in Eq. (10) drops out leaving one with $F \sim Ck_BT/au_I^2$. Combined with Eq. (6), this entails Eq. (1). Notably, this formula differs from Eq. (4.29) of Ref. [11], by a large logarithmic factor in the exponential. In view of this discrepancy (which has a conceptual importance, cf. the introduction), we present a rigorous derivation of Eq. (1) that clarifies the physics involved and yields the exact result $C = 8$ for the coefficient $C$.

In strong fields only the forward hops need to be considered [13,16], so that for the $i$th link of the optimal path we have $I = eP_{i+1,i}$ or, equivalently [cf. Equation (2)]

$$f_i (1 - f_{i+1}) \exp[u_i - (2/\alpha)|x_{i+1} - x_i|] = 1, \quad (12)$$

$$\epsilon_i \geq \epsilon_{i+1}, \quad \epsilon_i = \epsilon_i - F x_i. \quad (13)$$

A necessary condition for the existence of a physically acceptable solution, $0 < f_i < 1$, of Eq. (12) is

$$|x_{i+1} - x_i| < x_i = au_i/2 \quad (14)$$

(thus, the typical hopping length is field-dependent). A sufficient condition is known from the theory of a disordered asymmetric simple exclusion process [17], which is described by the identical equations. It is just a bit more stringent, $|x_{i+1} - x_i| \leq x_i - a \ln 2$. Since we are interested in the case $u_I \gg 1$, it is a legitimate approximation to assume that the condition (14) is both necessary and sufficient. Only the existence of the solution matters, not the actual values of $f_i$. (Those will be commented on at the end.) We expect on physical grounds that the optimal path does not deviate indefinitely from the equilibrium chemical potential, sup$|\epsilon_i| < \infty$. Then the standard percolation theory argument [13] entails

$$F = \min \lim_{i \to \infty} [(\epsilon_i - \epsilon_i)/(x_i - x_1)]. \quad (15)$$

Indeed, if $F$ were smaller than the right-hand side of this equation, then the contiguous path that obeys the constraints (13) and (14) would not exist. If $F$ were larger, the chosen path would be short-circuited by another one. To compute $F$ from Eq. (15) we use the fact that statistics of the random points $(x_i, \epsilon_i)$ is again an uncorrelated Poisson distribution of density $g$.

Consider the following lattice version of the problem. Let all coordinates be restricted to integer multiples of $b \ll x_j$. On every site $x_j = jb$ let there be a discrete set of energy levels $\epsilon_n(x_j), n \in \mathbb{Z}$, drawn from an independent Poisson distribution with the average energy spacing $1/\alpha b$. Suppose next that $(x_1, \epsilon_0(x_j))$ is the starting (the leftmost) site of the sought optimal path. Denote by $E(x)$ the largest $\epsilon_n(x)$ reachable from this site by a path that obeys the constraints (13) and (14), then $F = \lim_{\epsilon_{\alpha \to \infty}} \epsilon_\alpha(x_1) - E(x)/(x - x_1) = \lim_{\epsilon_{\alpha \to \infty}} E(x)/x$. Function $E(x)$ satisfies the obvious recurrence relation: $E(x)$ is equal to the largest $\epsilon_n(x)$ that still does not exceed $E(x)$, where $E(x) = \max E(y), x - x_j < y < x$. This relation is illustrated graphically in Fig. 2. It is easy to see that function $E(x)$ is steplike, i.e., that there is an increasing sequence of coordinates $x_j$, such that $E(x_j) = E(x_j) = \text{const}$ for $x_j < x \leq x_{j+1}$. Figure 2 aids in observation that given $x_j$, the next site in this sequence, $x_{j+1}$, is simply the one with the largest $E(x)$ less than $E(x_j)$ among all the points that belong to the vertical strip $x_i < y < x_i + x_j$. These $x_j$ are in fact the coordinates of the optimal path (more precisely, they become such once one further optimizes with respect

**FIG. 2.** Plots of $E(x)$ (thin line), $E_\alpha(x)$ (thick line) and the optimal path (dotted line). Ticks and dots represent lattice sites and their energy levels, respectively; $x_f = 9b$ is assumed.
to the path’s starting point). By virtue of this property, \( \Delta E_i = E_<(x_j) - E_<(x_{j+1}) \) and \( \Delta x_j = x_{j+1} - x_j \) are uncorrelated random numbers with average values \( 2k_B T_0/u_I \) and \( au_I/4 \), respectively, (in the limit \( b \ll x_j \)). This entails \( F = \langle \Delta E_i \rangle / \langle \Delta x_j \rangle = C k_B T_0/au_I^2 \) with \( C = 8 \).

It is worthwhile to clarify why Eq. (1) is not invalidated by inevitable large voids in the distribution of sites in the \((x, \varepsilon)\) plane. Whenever the optimal path meets such a void, \( \Delta E \) becomes much larger than its average value. However, the corresponding experimental contribution to \( F \) grows linearly with the size of the void whereas the probability of the void drops exponentially. These rare large voids have thus a negligible effect on \( F(u_I) \).

The derivation above indicates that the site occupations in the energy band \( |\varepsilon_j| \leq k_B T_0/u_I \sim (k_B T_0 Fa)^{1/2} \) that carries the current, deviate so much from their equilibrium values, \( \ln(1/f_j - 1) = e_j/k_B T \), that it plays no vital role in the transport. This agrees with the early result of Shklovskii [16] that at large \( |\varepsilon_j| \) the real temperature \( T \) in the last equation gets replaced by the “effective” one, \( T_{\text{eff}} = Fa/k_BT \). However, at \( e_j \)'s of relevance to VRH there exist large corrections, \( k_B T_{\text{eff}} \ln(1/f_j - 1) - e_j \sim (k_B T_0 Fa)^{1/2} \), to the asymptotic large-\( e \) behavior that prohibit one to derive Eq. (1) via a simple replacement \( T \to T_{\text{eff}} \) in the weak-field formula, Eq. (5).

Experimental implications.—The length \( L \) of real 1D conductors is seldom very large. Because of that their resistivity in a VRH regime fluctuates strongly from one sample to the next, depending on what configuration of breaks is realized [9,10]. Here we discuss only the ensemble average value of \( \ln \rho \). For the Ohmic case, in the common situation 1 \( \ll L \ll T_0/T \), where \( L = \ln(L/x_M) \), the result for this quantity is known to be [9,10] \( \langle \ln \rho(0, T) \rangle \propto \sqrt{2LT_0/T} \). In this regime practically all the applied voltage \( V = FL/e \) drops on the single largest break [10]. Hence, the Ohmic behavior lasts as long as \( eV < k_BT \). In some range of \( eV \) above \( k_BT \), the same break remains the bottleneck for the transport. Using Eq. (3) and averaging over the position of the break along the chain, we obtain the exponential dependence,

\[
\ln(\rho/\rho(0, T)) = -eV/2k_BT, \quad k_BT \ll eV \ll eV_L.
\] (16)

The upper limit \( V_L \) where this formula is valid can be estimated from the condition \( eV_L \sim \langle \Delta \xi \rangle \sim k_BT_0/u_I \), which gives \( eV_L \sim k_BT_0/L^{3/2} \). At \( V \gg V_L \), the infinite-chain results, Eqs. (11) and (1), apply.

On a qualitative level, existing experimental data are consistent with our theory. For example, a characteristic \( S \)-shaped \( \log I/\log V \) curve [4] follows naturally from our equations. To unambiguously verify them an earnest effort and a thoughtful experimental design are required. At present, our knowledge and degree of control over disorder and other parameters of 1D systems remain poor; therefore, a wide dynamical range of data is needed to compensate for many uncertainties. The most suitable for this purpose are systems where localization length is not too short, e.g., high-quality carbon nanotubes. Their resistance has to be measured over a broad range of low \( T \), where \( \rho \) is determined by the exponential terms we computed rather than some power-law prefactors we left undetermined here. Care should be taken to remain in the validity domain of the theory, \( L \gg x_M \sim a\sqrt{T_0/T} \). The parasitic effects of contact resistance [3,5] need to be done away with reliably. If the system is quasi-1D rather than one-dimensional, hopping across the chains [4,5] must be negligible. Finally, a careful statistical analysis of the data is likely to be needed [18].

An important qualitative prediction of our theory is that enormous sample-to-sample variability of the low-\( T \) hopping resistance becomes less of an issue in the non-Ohmic regime. This may facilitate both the experimental studies we call for and the future theoretical work that would be needed to include the effects of Coulomb interaction into the model. Such coupled experimental and theoretical studies will likely to shed much light on the intriguing transport properties of low-dimensional conductors.

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[14] In Ref. [10] only symmetric diamonds were considered.
[15] The calculation of the prefactor is a difficult open problem [2] while the result is likely to be nonuniversal.