LOW-TEMPERATURE VRH CONDUCTION THROUGH COMPLEX MATERIALS IN THE PRESENCE OF A TEMPERATURE-DEPENDENT VOLTAGE THRESHOLD: 
A SEMI-CLASSICAL PERCOLATIVE APPROACH

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Abstract

In this paper, we study the variation of low temperature ($T$) $dc$ conductance, $G(T)$, of a semi-classical percolative Random Resistor cum Tunneling-bond Network (RRTN), in the presence of a linearly temperature-dependent microscopic voltage threshold, $v_g(T)$. This model (proposed by our group in the early 90’s) considers a phenomenological semi-classical tunneling (or, hopping through a barrier) process. Just as in our previous constant-$v_g$ case, we find in the present study also that the variable range hopping (VRH) exponent $\gamma$ varies continuously with the ohmic concentration $p$ in a non-monotonic fashion. In addition, we observe a new shoulder-like behaviour of $G(T)$ in the intermediate temperature range, below the conductance maximum.

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1 Introduction

We study the dc electrical conductance, $G(T)$, at a low temperature ($T$), such that the thermal energy $k_B T$ ($k_B$, being the Boltzmann constant) is small compared to either the disorder energy of the non-interacting carriers or the Coulomb interaction energy between pairs of spinless charge carriers in pure systems. The topic has drawn enough attention among the scientists for the last several decades and continues to draw interest. For binary (metal-insulator) composites, which are insulating at zero-temperature, it may be quite hard to conduct electricity at sufficiently low temperatures with the standard diffusion process only. But with the help of thermal energy, a localised fermion (electron or hole) may be able to hop from one site to another. The conduction of the spinless and chargeless fermions through the disordered lattice, with the help of a phonon-assisted hopping process, can be quantitatively described by Mott’s Variable Range Hopping (VRH) formula [1] as,

$$G(T) = G_0 \exp \left[- \frac{T_0}{T} \gamma \right],$$

where $\gamma = 1/(d + 1)$ for a $d$-dimensional sample (i.e., $\gamma = 0.25$ for a 3D sample). The $T_0$ is a sample-specific characteristic temperature scale, above which the conduction sample-specific characteristic temperature scale, above which the conduction is due mainly to the phonon-scattering of the charge carriers (i.e., electrons/holes). Further below $T_0$, there is another sample-specific temperature $T_m$, where the bulk resistance due to coherent/quantum fluctuation matches the resistance due to incoherent/thermal fluctuations and the combined resistance (conductance) goes through a minimum (maximum) as a function of temperature, $T$. In a quantum insulator, for a temperature, $T < T_m$, the coherent tunneling effect dominates even further, while the incoherent scattering due to the phonons (hence, the ohmic resistance) decreases. We are interested in the low-temperature behaviour ($T << T_0$ or, $T_m$) of the resistance due mostly to the coherent tunneling of the carriers and very little to the incoherent scattering due to phonons.

In a pure system (i.e., without disorder), where the localisation may be due to the effects of the Coulomb interaction between the spinless fermions, Efros and Shklovskii [2] found that $\gamma = 0.5$ in Eq.(1) for a sample in any dimension. Further, if such interacting systems possess percolative disorder, clusters and connectivities would play important roles in the conduction processes, and may be experimentally observed in many nano-structured materials.

A phenomenological percolation-like picture of this quantum transport of an insulating ($T = 0$) sample, at a low but finite temperature, was discussed by Ambegaokar, Halperin and Langer [1]. The charge transport in a macroscopic system, with no system-spanning cluster, occurs mainly due to the hopping between nearby finite-sized microscopic/macroscopic metallic clusters (a classical concept), which may be considered as the spatial extent of isolated localised states (a quantum concept) and the possibility of tunneling between between some of them. Mi-
croscopically, we achieve it in a model by introducing the possibility of a semi-classical tunneling (or, a classical dielectric breakdown) through the insulating bond between two nearest neighbour (i.e., separated by a single bond-length only, and no farther, for simplicity) ohmic-bonds in an underlying Random Resistor Network (RRN). These special insulating bonds are called tunneling or t-bonds. A t-bond becomes active (i.e., permit tunneling) when the magnitude of the voltage-difference, $v$ across it is larger than some pre-assigned microscopic threshold voltage, $v_g$, i.e., when $v \geq v_g$. One can imagine a non-percolating RRN in such a situation to percolate through the intermediary of some t-bonds at the proper places. It is also clear that the network would need a macroscopic threshold voltage $V_g$ to make the crucially connecting t-bonds active. Historically [3], such a network has been named a Random Resistor cum Tunneling-bond Network (RRTN).

Thus this percolative RRTN model does simultaneously possess disorder as well as Coulomb interaction effects; the latter in the form of a voltage threshold or barrier, $v_g$ (cf. Coulomb blockade between quantum dots) across the t-bonds. They give rise to an implicit microscopic nonlinear $i$-$v$ characteristic. Within a RRN type classical bond percolation model, one may consider the transport process as purely diffusive through the randomly placed ohmic bonds (or, o-bonds). But, in our RRTN model, a pair of RRN-wise disconnected metallic clusters (or, localized states) may become linked by the hopping or tunneling process. A single t-bond may complete the connection between two nearby distinct clusters and the distance between their centres of "mass" may vary widely in different parts or realizations of the percolative network. Since this effective hopping covers various possible lattice distances in the RRTN, it describes the VRH phenomenon quite well [4].

Further, as proposed in the works of the Refs. [6, 7], we find a generalization of the original Mott’s VRH formula with a power-law prefactor, namely,

$$G(T) = G_0 \left( \frac{T_0}{T} \right)^s \exp \left[ - \left( \frac{T_0}{T} \right)^\gamma \right], \quad (2)$$

to be the needful for both our previous [4] as well as the present works on the VRH. Quite generally, it is more appropriate for application on a wide variety of (composite to nano-) materials, where the diffusive and the activated transport processes compete with each other and dominate over different temperature regimes. But, whereas the microscopic voltage threshold $v_g$ was a constant independent of the ambient temperature $T$ in our previous works [4], this microscopic gap voltage $v_g(T)$ is allowed a linear $T$-dependence, and we study its effect in this paper. Our study was motivated by an experiment [5] on single wall carbon nanotubes (SWNTs). The rest of this work is organized as follows. In the section 2, we describe the thermal version of the RRTN model and the numerical solution of the electrical equation of continuity. The resulting conductances for different sizes $L$ of the network are sieved through finite size scaling analysis to obtain the exponents $\gamma$ and $s$ at low $T$ for various $p$’s. In the section 3, we discuss the results and end with some concluding remarks.
2 Numerical works

As stated above, we study the effect of a linear $T$-dependence of the gap energy, $\Delta(T) = qv_g(T) = qv_g(0) - k_BT$, required for the passage of a charge carrier through a t-bond, on the VRH exponents. Here, $q$ is the effective charge of each carrier and $v_g(T)$ is the effective threshold or gap voltage of a t-bond at the temperature $T$. Thus, the zero-$T$ gap energy is $\Delta(0) = qv_g(0)$, where $v_g(0)$ is the $T$-independent $v_g$ of our previous works [3, 4]. Clearly, $v_g$ plays the role of the gap between the conduction band and the valence band of a semiconductor. As no percolation model explicitly contains temperature, we have parametrized a couple of simple but logical temperature-dependence for the microscopic ohmic and the tunneling bonds of the RR TN model, symbolised as $g_o(T)$ and $g_t(T)$ respectively, in our work. In particular, we choose the functional form of a dirty metal for the $o$-bonds, i.e., $g_o(T) = 1/(r' + a_0T)$; and a thermally activated form for the $t$-bonds, namely, $g_t = b \exp\left[-\Delta(T)/k_BT\right]$ for the $t$-bonds. We may also write $g_t = b \exp\left[-v_g(T)/k_eT\right]$, where $v_g(T) = v_g(0) - k_eT$ and $k_e = k_B/q$.

The values of the parameters chosen are: the microscopic residual (impurity effect) resistance of the $o$-bonds $r' = 1$, the thermal coefficient of resistance $a_0 = 0.01$, and $k_e = 0.005$. Further, the zero-temperature voltage threshold of the $o$-bonds $v_g(0) = v_g = 0.5$ and $b = 10$, as in our previous works on VRH. Also, since an insulating bond has zero conductance, it has no variation with $T$. Further, the gap energy cannot be negative (unphysical), and $v_g(T) = v_g(0) - k_eT \geq 0$. This defines a temperature, $T_m = v_g(0)/k_e$, upto which the linear relation for $v_g(T)$ is valid and $v_g(T \geq T_m) = 0$. In the units in which $q = 1$, which we have chosen in all our previous works, $k_e = k_B$. So for this choice of parameters, $T_m = 100$ and $g_t = 10 \exp[-100/T]$. For $T \geq T_m$, each realization of an RRTN behaves like its corresponding maximal RRTN configuration, as all the geometrically allowed t-bonds are active. Clearly, $T_m$ is the exact temperature at which the macroscopic conductance reaches its maximum value $G_m$.

For obtaining both the steady state ($dc$) and the dynamic ($ac$) properties, we have used in all our works [3, 4, 8, 9] beginning with the earliest one, the method which may be called lattice Kirchhoff’s dynamics discussed explicitly in Ref.[9]. In passing, we mention that recently we have studied this dynamics in the RRTN model for comparison with the non-equilibrium dynamics of various laboratory experiments and of natural occurrences [8, 9]. We point out that in the absence of joule heating effect (which is not our primary concern in the study of VRH, and which is achieved experimentally by using a good thermostat), there are three $dc$ regimes: (i) a lower linear (ohmic) regime for $V < v_g$ (ii) an upper linear regime (ULR) for $V >> v_g$, and (iii) an intermediate sigmoidal regime for $V \geq v_g$. There is no need to study VRH in the lower regime since it belongs to the RRN and there is no VRH. In the ohmic ULR, all the $t$-bonds are active irrespective of the value of $v_g$, since the driving voltage is very high. But, in the sigmoidal regime (iii), only some of the $t$-bonds are active in a rather complicated fashion, depending on the configuration of the RRTN under a medium voltage. The case of ULR has been discussed
Conductance, $G(T)$

Temperature ($T$)

$L=80$, $V=20$, $p=0.20$

$p=0.30$

$p=0.40$

$p=0.50$

$p=0.60$

$p=0.70$

Figure 1: The low temperature macroscopic conductance $G(T)$ versus $T$ for some RRTN samples with $L = 80$ and different $p$'s ($0.2 - 0.7$), under an external bias of $V = 20$ volt. This figure shows a nonmonotonic decay followed by a rise of the $G(T)$ at lower $T$'s, in the percolating samples ($p \geq 0.5$). Interestingly, these graphs show a shoulder-like behaviour (near but below $T = T_m$) which were completely absent in the cases with $T$-independent $v_g$ [4].

(for a constant $v_g$) elsewhere [4]. We focus here on the sigmoidal regime.

In this study of VRH in the sigmoidal regime, more t-bonds get activated with the rise of not only the driving voltage $V$ but also the temperature $T$, since $v_g(T)$ decreases with $T$. So, we kept the voltage across the macroscopic sample fixed at a value of $V = 20$, where some but not all the allowed t-bonds are conducting. This fact disallows any reasonable mean-field study (e.g., the effective medium approximation or EMA) of this situation as they are devised to calculate an average conductance in the maximal RRTN limit (i.e., the ULR). In our numerical works, we calculate the differential conductance $G(T) = dI/dV$ at various values of $T$, for the system sizes $L = 20 - 100$ and the ohmic concentrations $p = 0.2 - 0.7$. In the Fig. 1, for example, we show $G(T)$ versus $T$ for some $L = 80$ samples with these $p$'s and a bias of $V = 20V$. We note that the conductance maxima $G_m$ occur, as before [4], around a $T_m$ of about 85 (in our arbitrarily chosen units) for different choices of $L$, $p$ and the material parameters $a_0$, $b$, $u$ and $r'$. The salient feature of these curves (different from our works with a constant $v_g$) is that for each of them, there is a shoulder-like region in $G(T)$ below the conductance maximum, and around a $T = 60$.

For the RRTN samples with $p \geq 0.5$ (as $p_c = 0.5$ for the 2D square lattice RRN), we have observed a non-monotonic sharp rise and then an eventual saturation in the $G(T)$ to a finite residual value, at sufficiently low temperatures. This originates from the residual value in $g_0(T)$ (i.e., $r'$) for $T \to 0^+$; thus, the finite value for $G(T \to 0^+)$ depends on the choice of the parameter values during the numerical calculation. These configurations with $p \geq 0.5$, are
The excess macroscopic conductance, calculated from the Fig. 1, by subtracting the RRN contribution at each $T$ from the corresponding $G(T)$ of the RRTN. These graphs also retain the shoulders as observed in the Fig. 1. This indicates that these anomalous shoulder contributions are due to the new paths in the RRTN through the extra tunneling bonds, whose contributions increase with temperature as the $v_g(T)$ decreases linearly with it.

all of RRN-wise percolative type with system-spanning ohmic backbone. For any finite system size, it is probable to get a RRN-wise percolating sample for almost any $p$. But for $p \geq p_c$, this possibility does increase with increasing $p$. The possibility of getting a percolating sample at some $p > p_c$, increases with the rise in the system size $L$ and, for $p < p_c$, this possibility decreases with increase of $L$. This is due to the fact that, for infinite-sized samples, each of them is percolating over $p_c$. Such an observation is qualitatively very similar to several other reports like, Refs. [4, 11, 12] etc.

We subtract the RRN conductance at each $T$ from that of the corresponding RRTN conductance to calculate the excess-conductance, $G(\text{RRTN}; T) - G(\text{RRN}; T)$, for each sample. This was introduced in our previous work [4] to bring a classically percolating and a classically insulating sample on an equivalent footing since VRH, after all, is a property of insulators. In the Fig. 2, we have shown the excess conductances for the same sets of RRTN samples as in the Fig. 1. The shoulder-like behaviour exists similarly in these graphs as well. This behaviour is related to the choice of a linear reduction of the gap voltage for the t-bonds with increase in temperature. Since we are looking for a scaling property of VRH in the RRTN, we have used our previous experience in the absence of anything better. Unfortunately, we find that a similar scaling with the normalised variables excess $[G/G_m]$ versus $T/T_m$, as we had used in the Ref. [4] for the case of $T$-independent $v_g$, does not work so well with $T$-dependent $v_g$, as revealed by the Fig. 3.

For fitting our numerical data with Eq. (2), we have adopted a careful nonlinear fitting scheme, based on simplex method, which is independent of the derivative values (Jacobian) of the minimization functional, for all of our numerical results. As before, we find a strong non-self-
averaging behaviour among the steady-state conductance values for different samples with same low $p$'s and any $L$. Thus we worked with one sample at a time and analysed its results. But as a very common practice in statistical mechanics, we find the exponents in the finite-sized samples (one particular RRTN sample of each $L$), and then use those values to calculate the value in the thermodynamic limit by an appropriate finite-size scaling analysis (FSA). Under this procedure, any exponent for a particular $L$ becomes very much sample-specific and differ appreciably for different ensemble members with the same $L$ and $p$. To escape from this difficulty, we scale our numerical data with the corresponding maximum values, and use the normalized data, i.e., excess $G/G_m$ vs. $T/T_m$, during the nonlinear fitting. In the FSA, we use the relation $\gamma(L \to \infty)$ as, $\gamma(L) = \gamma(L = \infty) + A L^{-\nu}$, and find out that in all the samples the best-fit is achieved with a $\nu \simeq 1$.

As the nonlinearity is very strong in the sigmoidal regime of the dc characteristics, the variable range hopping phenomenon dominates the conduction process. So, for this work we chose the dc regime where the effect of $v_g(T)$ is most prominent. In general, to test the quality of the generalised VRH formula, Eq.(2), in describing the $G(T)$ versus $T$ data, we have formed four non-overlapping regimes of reduced temperatures $t \equiv T/T_m$, namely (i) $1.0 > t \geq 0.75$, (ii) $0.75 > t \geq 0.50$, (iii) $0.50 > t \geq 0.25$, and (iv) $0.25 > t \geq 0.10$. It may be noted here that it is very difficult numerically to get reliable values of $G(T)$ for $t < 0.1$, when the dopant concentration $p$ is close to (e.g., $0.2 - 0.3$ in our 2D square lattice case) to the corresponding tunneling percolation threshold $p_{ct}$ (0.18 for a square lattice [3]), and when the driving voltage is close to the system’s threshold $V_g$.

For calculating the exponents (i.e., $\gamma$ and $s$), we have always used the excess-conductance for

Figure 3: A scaling plot with the variables: normalized excess $(G/G_m)$ versus $T/T_m$, for different system sizes $(L)$ with $p = 0.3$ under $V = 20V$. This scaling was motivated by our previous work [4], where it works out quite nicely.
Figure 4: The variation of the VRH-exponent $\gamma$ with different $p$-values for four different $\Delta t$ (where scaled temperature, $t \equiv T/T_m$) intervals, as described in the section on numerical works. Each vertical line indicates a fixed $p$. Each zig-zag shaped curve joined by straight lines through two consecutive $\gamma p$-values for a given $\Delta t$ interval, is only for the guidance of the eyes. These graphs indicate an asymmetric, non-monotonic behaviour with $p$.

all the $p$ values because only the excess-conductance data for all $p$'s have a qualitatively similar behaviour with the Eq. (2). The VRH exponents extracted from the $G(T)$ data for each of the above $\Delta t$ intervals are calculated for all the system sizes ($L = 20 - 100$) at each $p$ (0.2 - 0.7) separately. Then the finite size effects in the exponents were removed using the FSA. We observe that the values of the exponents for each $p$ depend on the choice of the temperature-bins $\Delta t$. We find that the behaviour of $\gamma(p)$ is non-monotonic with $p$ and it is quite asymmetric around the $p$ values. This behaviour of $\gamma(p)$ versus $p$ is shown for the above RRTN samples in the Fig. 4. The exponent $\gamma$ first decreases with the lowering of $T$, and then from some low enough $T$ (near $T/T_m = 0.18$), it increases again. The graphs in this figure are devised to emphasize the need to extrapolate the obtained exponents to much lower temperatures, not accessible directly.

We notice from our numerical results that the calculated values of the more important VRH exponent $\gamma$ is almost always greater than 0.5. The estimated values of the other VRH exponent $s$ always lie close to 2, similar to the comment by Aharony et al. [6].

3 Discussions

From a very low dilution ($p$), as the system is further diluted (or, disordered), the resistor elements corresponding to the ohmic and the tunneling bonds, combine in a complex fashion to form the tenuous current carrying paths of the network. It results in a nontrivial, non-monotonic conductance as a function of temperature, $T$. Due to the empirical temperature dependences of the microscopic conductances, $g_o(T)$ decreases and $g_t(T)$ increases, with the rise in $T$. When the
collective current contributions from both the o- and t-bonds are comparable (thus the exact occurrence depends on a particular RRTN bond-configuration), the macroscopic differential conductance attains a peak value of $G_m$ at $T = T_m$. In general, the RRTN with larger $p$’s are more conducting, at least, at high enough $T$’s as it contains more conducting (ohmic) paths. For a RRN non-percolating lattice, there is no system spanning geometrical connectivity through the o-bonds only. Thus for getting a system spanning conducting channel in the full macroscopic RRTN, several t-bonds at some appropriate gap (insulating) sites have to become active in the transport process. Now, due to our consideration of pure activated behavior of each t-bond, the conductance $g_t$ for each of them, is decreasing exponentially with the inverse-$T$ argument. So, they are highly resistive in the low-$T$ regime. In contrast, the o-bonds behave like dirty metals (due to the presence of microscopic residual resistance, $r_0 \equiv g_0^{-1}$) and, hence apart from the saturation value of $g_0$ at $T = 0$, $g_o$ diverges as an inverse power as $T \rightarrow 0$. As a t-bond appears necessarily in the gap between two next-nearest neighbor o-bonds, so under these behaviours of $g_o$ and $g_t$ with temperature, these semi-classical paths across t-bonds are almost blocked due to their very high resistance. Thus the macroscopic conductance follows a similar variation as a t-bond at low $T$, since the exponential function has a faster gradient than a power-law function. When the conductance of the RRN at some $T$ (obviously, not of all the o-bonds separately) is subtracted out from the RRTN conductance, the contribution of the o-bonds remain through the power-law prefactor with an almost constant exponent, $s$. For the same reason, the exponent $\gamma$ in the low-$T$ regime is close to unity. Moreover, as we are investigating a system, consisting of both metallic (diffusive) and activated (hopping) processes, it is quite reasonable to consider a mathematical formula (like Eq. (2)) that contain both the terms, i.e., a power-law function and an exponential function, of $1/T$. In this regard, the generalised-VRH formula (i.e., Eq.(2)) works better than the Mott’s VRH formula (i.e., Eq.(1)), which contains only an exponential term.

At sufficiently high temperatures, i.e., $T \rightarrow \infty$ (or, practically speaking, when $T \gg T_0$), a purely metallic behaviour appears in $G(T)$ for all RRTN samples of any $p$. But for the limit $T \rightarrow 0$, the explicit saturating tendency in the microscopic temperature dependence for the $g_o$, gets only reflected in the bulk $G(T)$ of those RRTN samples, whose RRN is percolating. For a RRN percolating sample, a significant portion of the bulk current through the corresponding RRTN is due to the ohmic backbone. Thus at low-$T$’s, where all the parallel paths are blocked by the highly resistive t-bonds, only the RRN channels take care of the whole conduction through that RRTN. Thus the macroscopic $G(T)$ for those temperatures is dominated by the respective temperature dependences of the ohmic bonds, i.e., by $g_o(T)$. Thus in these samples, we find the proper signature of the VRH only in the excess conductance, where the RRN contribution has been subtracted out from the RRTN conductance.

To conclude, our VRH study belongs to the class of the non-Mott, non-Efros-Shklovskii type. Further, we find a continuous variation of the exponent $\gamma$ with $p$. Throughout the paper, we have
given more emphasis on the behaviour of $\gamma$ than on $s$, because we are basically interested in the low-$T$ domain, where the exponent $s$ has been observed to be quite robust. Stated differently, the relative variation of $s$ is almost insignificant compared to that of $\gamma$ for these temperatures. Further, in the case of a $T$-dependent threshold voltage, we find a shoulder-like behaviour in $G(T)$ below its maximum ($T < T_m$). This flat regimes indicate the existence of some smaller peaks in $G(T)$ at some $T < T_m$. For quite high temperatures ($T >> T_m$, near corresponding $T_0$), we achieve a purely metallic behaviour, i.e., the exponents are $s \sim 1.0$ with $\gamma \rightarrow 0^+$. In a purely quantum-mechanical description, this regime is attributed to a completely decoherent regime of the electronic wave-function due to inelastic-scattering with the phonons. From the property of the global maximum of $G(T)$, one may easily find that the $T_0$ and the $T_m$ are mathematically related by the expression, $T_0 = T_m \left(\frac{s}{\gamma}\right)^{1/\gamma}$. In our previous work [4], we had noted that $T_0$ is a temperature (energy) scale which augura the onset of almost diffusive behavior (i.e, $s = 1$), and

with that interpretation we find in this work that $T_0 = T_m$.

References


