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General resistance crossover expressions for three-dimensional variable-range hopping

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Abstract. We observe a crossover in the temperature dependence of the variable-range-hopping resistivity in a three-dimensional nickel–silicon film from the Mott $T^{-1/4}$ -behaviour to the soft-gap $T^{-\nu}$ -behaviour with $\nu \approx 0.72$. We propose general expressions for describing such crossovers from $T^{-1/4}$ -behaviour to $T^{-\nu}$ -behaviour for any ν from $1/4$ to 1 . The theoretical expressions fit the experimental data well.

1. Introduction

Crossovers in the temperature dependence of the three-dimensional (3D) variable-range-hopping (VRH) resistance $R(T)$ from the Mott behaviour [1]

$$R(T) = R_0 \exp(T_M/T)^{1/4} \quad T_M = \beta_M/k_B G_0 \xi^3 \quad (1)$$

at high temperatures to the Efros–Shklovskii (ES) behaviour [2, 3]

$$R(T) = R_0 \exp(T_{ES}/T)^{1/2} \quad T_{ES} = \beta_{ES} e^2 / k_B \kappa \xi \quad (2)$$

at low temperatures have been observed in various materials [3–8]. The Mott $T^{-1/4}$ -law of equation (1) was obtained by assuming the localized density of states (DOS) to be constant near the Fermi level: $G(E) \equiv G_0 = \text{constant}$. The ES $T^{-1/2}$ -law of equation (2) is a consequence of the Coulomb gap (CG):

$$G(E) = \alpha_3 E^2 \quad \text{with } \alpha_3 = (3/\pi)(\kappa^3/e^6) \quad (3)$$

which is opened up in the DOS at the Fermi level as a result of the Coulomb interaction between localized electrons. Here ξ is the localization length, e is the elementary charge, and κ is the dielectric constant. The one-particle energy E is measured from the Fermi level.

Using the effective 3D DOS of the form

$$G(E) = \alpha_3 E_{cg}^2 E^2 / (E_{cg}^2 + E^2) \quad (4)$$

which approaches the Mott constant DOS $G_0 \equiv \alpha_3 E_{cg}^2$ in the limit of large E and which approaches the parabolic DOS of equation (3) in the opposite limit of small E , we derived expressions which describe quite well the experimental resistance data for the crossover from the Mott $T^{-1/4}$ -law to the ES $T^{-1/2}$ -law, as observed in amorphous $\text{Ni}_x\text{Si}_{1-x}$ and In_xO_y films [8]. Other theories of the Mott–ES crossover, given by Aharony *et al* [9] and by Meir [10], are also claimed to be in good agreement with experiments. It should be

noted that all of these theories, aiming to describe the crossover from the Mott law at high temperatures to the ES law at low temperatures, are based on the concept of the CG of equation (3).

The CG and its manifestation of the ES $T^{-1/2}$ -law were the controversial subjects of a number of critical reviews [11–13]. A parabolic gap in the DOS at the Fermi level is undoubtedly recognized in many current computer simulations [2, 3, 14–17]. The ES law of equation (2) is certainly observed to be followed in various materials [2–11]. Massey and Lee [18, 19] also reported a direct observation of the parabolic gap in boron-doped silicon by tunnelling spectroscopy. However, in spite of all of these achievements, the problems of the CG and of the ES $T^{-1/2}$ -law are still under discussion [11–13].

In fact, both computer simulations and experimental measurements have been unable to quantitatively identify the form of the DOS in the limit of low energies. While all of the computer simulation data show a parabolic DOS at not very low energies, there is a considerable quantitative discrepancy between the forms of the DOS at very low energies. An existence of a gap harder than that of equation (3) had earlier been argued for by Efros [20] and by Davies *et al* [14]. Instead of the square of the energy in equation (3), Mobius *et al* [15] suggested a power of approximately 2.6, and Sarvestani *et al* [16] suggested a similar power of 2.7. Theoretical arguments incorporating at least some classes of many-electron correlated transitions always lead to a functional dependence of $G(E)$ stronger than the parabolic form of the CG of equation (3) at very low energies [13].

Experimentally, besides the above-mentioned Mott–ES crossover, there are two classes of crossovers from the Mott $T^{-1/4}$ -VRH to the $T^{-\nu}$ -VRH with the exponent ν considerably greater than 1/2. In the first class, the exponent ν firmly tends to 1 in the limit of low temperatures [21–23]. Such an activation T^{-1} -behaviour of $R(T)$ is generally accepted as a manifestation of the so-called hard gap (a magnetic gap [21, 22] or non-magnetic gap [23]) in the DOS at the Fermi level. A typical feature of these data is that the resistance $R(T)$ follows the Mott $T^{-1/4}$ -law well at first and then the ES $T^{-1/2}$ -law over sufficiently large temperature ranges, before abruptly changing to the activated T^{-1} -behaviour at very low temperatures. In the second class of crossovers, there is no clear picture of the three VRH regimes; the exponent of the resistance smoothly increases from the Mott value of 1/4 to values which, although greater than 1/2, are still considerably smaller than 1. Such values of the general VRH exponent ν at the low-temperature limit should be considered to be related to soft gaps that are ‘harder’ than the CG at the Fermi level. Very narrow gaps deduced from VRH data were recently reported by Zabrodskii and Andreev [24] and by Lee and Massey [19].

In this paper, we present data on the smooth crossover in the temperature dependence of the VRH resistivity from the 3D Mott behaviour to the soft-gap VRH regime, where the VRH exponent ν is approximately 0.72. We then propose general expressions for the crossovers from $T^{-1/4}$ -behaviour to $T^{-\nu}$ -behaviour with any ν ranging from 1/4 to 1. The theoretical expressions describe the data well.

2. Film preparation

Thin amorphous $\text{Ni}_x\text{Si}_{1-x}$ films were fabricated by co-evaporating Ni and Si using two electron guns. Seven to eight narrow glass slices of 2.5 mm width were glued onto a microscope glass slide. These narrow glass segments were employed to avoid shadowing problems, which are introduced when using a mask. Three microscope glass slides were placed ‘end to end’ above and between the Ni and Si graphite boats in order to obtain the differences in Ni content between the samples. Small broken glass pieces covered with

photoresist were glued between these narrow glass slices, so that EDAX (energy-dispersive analysis of x-rays) samples would also be available. Typical evaporation rates were 0.7 \AA s^{-1} and 11.5 \AA s^{-1} for the Ni and Si sources respectively. The evaporations were carried out in a vacuum of 10^{-6} mm Hg, and the glass slices were held at room temperature to avoid crystallization of the amorphous Si. The Ni and Si have purities of 99.9% and of 99.97% respectively. As a check against contamination, the original graphite boats were replaced with new boats containing new charges. The data were reproducible from series to series. None of the series exhibited superconducting properties, which could possibly arise from boat contamination, particularly from Cu.

The homogeneity of each film is very important near the metal–insulator transition. We had experienced great difficulty in stabilizing the Si evaporation rate, resulting in very inhomogeneous films. However, we noticed that the Si evaporation rate would generally stabilize to a steady value if the Si boat was heated for a least ten minutes. Quartz crystal monitors, positioned above each graphite boat, monitored the evaporation rates and hence the thicknesses of the Ni and Si films deposited during the evaporation. The Ni thickness was then plotted versus the Si thickness; any deviation from a straight line indicated instability in the evaporation rate of one of the two materials. Thus, any series that exhibited deviations from a straight-line fit was considered to be inhomogeneous and was discarded. Numerous evaporations were attempted before an insulating homogeneous series was obtained. We believe that this series is homogeneous to better than $\pm 2.5\%$ throughout the typical thickness of 900 \AA . The film thickness was measured using the Tencor Instruments ‘Alpha-step’ 200 instrument.

The Ni content for the film being investigated is estimated at 5.4 at.% Ni from the EDAX and Rutherford back-scattering data. Since the metal–insulator transition occurs at around 20 at.%, this film is very strongly insulating. The geometric factor f_g , used to convert resistance to resistivity, is 1.3×10^{-6} cm. Thus, since the room temperature resistance of this film was $27\,600 \text{ \Omega}$, its room temperature resistivity would be 0.038 \Omega cm or its room temperature conductivity would be $27 \text{ \Omega}^{-1} \text{ cm}^{-1}$. The Keithley 617 electrometer was used to measure the film resistance. Standard low-temperature liquid helium cryostats were employed.

3. Experimental results

The VRH exponent ν and the characteristic temperature T_0 in the general VRH resistance expression $R(T) = R_0 \exp(T_0/T)^\nu$ can be simply determined from the R versus T data using the well-known technique of Hill [25] and of Zabrodskii and Zinov’eva [26]. These authors suggested calculating the quantity $\omega(T) = -d \ln R / d \ln T = \nu(T_0/T)^\nu$ from the resistance data $R(T)$, and then making a linear regression fit to the $\log \omega(T)$ versus $\log T$ data. The slope of the linear fit is equal to the VRH exponent ν , and the intercept I of the linear fit is related to the characteristic temperature T_0 via the expression $T_0 = (10^I / \nu)^{1/\nu}$. Thus, one can readily determine whether the resistance data follow the Mott $T^{-1/4}$ -law or the ES $T^{-1/2}$ -law or another general $T^{-\nu}$ -VRH dependence.

The resistance data for the amorphous $\text{Ni}_x\text{Si}_{1-x}$ film are presented in figure 1. As is seen there, this film does indeed exhibit the 3D Mott VRH $R(T)$ behaviour at high temperatures, where the expression

$$R_M(T)/M\Omega = 0.00283 \exp(7310/T)^{0.27}$$

fits the data very well. Here the Mott characteristic temperature $T_M = 7310 \text{ K} \pm 300 \text{ K}$ and the VRH exponent $\nu = 0.27 \pm 0.02$. However, in the liquid helium temperature region all

3D Mott to VRH Resistance Data for an Amorphous Nickel-Silicon Film

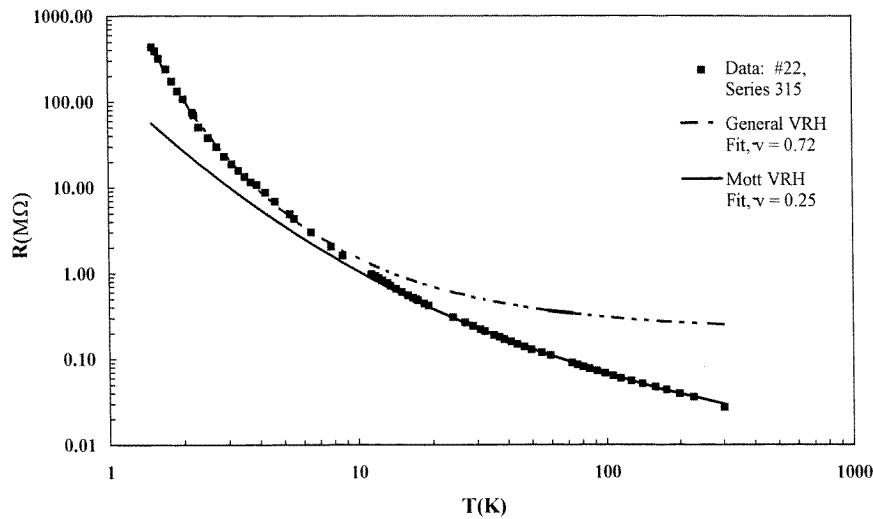


Figure 1. Resistance versus temperature for a 900 Å amorphous $\text{Ni}_x\text{Si}_{1-x}$ film having 5.4 at.% Ni. The high-temperature data can be fitted nicely using a Mott 3D VRH law. The low-temperature data can be fitted using a general VRH law that has a VRH exponent of 0.72, which is considerably greater than the ES VRH exponent of 1/2 and considerably smaller than the hard-gap exponent of 1.

of the data certainly tend to follow the general VRH law $\ln R(T) \propto T^{-0.72}$ rather than the expected ES law $\ln R(T) \propto T^{-0.5}$. The expression

$$R_{VRH}(T)/M\Omega = 0.215 \exp(25.2/T)^{0.72}$$

seems to fit the low-temperature data quite well, as is observed in figure 1. The characteristic temperature T_0 of this general VRH regime is $25.2 \text{ K} \pm 2 \text{ K}$ and the VRH exponent $\nu = 0.72 \pm 0.03$. Note the smoothness of the crossover between the two limits. From figure 1 the crossover temperature T_c is also estimated; the value is found to be $9 \text{ K} \pm 3 \text{ K}$.

4. Crossover expressions and comparison with the data

We now derive expressions for the general crossover from the 3D Mott $T^{-1/4}$ -VRH law to the general $T^{-\nu}$ -VRH law for any ν in the range $1/4 \leq \nu < 1$. These expressions include the 3D Mott-ES crossover theory of reference [8] as a special case.

The observed smooth resistivity crossovers should also be reflected in the expressions for the DOS, encompassing the Mott constant DOS as well as the ‘harder’ soft-gap DOS where $G(E) \propto |E|^n$ with $n > 2$. Following Hamilton [27], the power n of the energy in the 3D DOS is related to the VRH exponent ν in the 3D VRH resistivity by the expression

$$\nu = (n + 1)/(n + 4) \quad n \geq 0. \quad (5)$$

Therefore, to describe crossovers from the 3D Mott $T^{-1/4}$ -VRH to the $T^{-\nu}$ -VRH, in analogy with the 3D Mott-ES crossover theory of reference [8], and with our theory for general

$T^{-1/3}$ -to- $T^{-\nu}$ crossovers in two-dimensional VRH [28, 29], one can choose a ‘universal’ 3D DOS in the form

$$G(E) = \alpha E_{sg}^n |E|^n / (E_{sg}^n + |E|^n) \quad (6)$$

where the power n of the energy can be related to the VRH exponent ν using equation (5), and where α and E_{sg} depend upon n and are constants for a given n . For the case of $n = 2$, this DOS coincides with the DOS of equation (4), used for describing the Mott–ES crossover in reference [8].

On the basis of the DOS of equation (6), using the Mott optimizing procedure (see, for example, references [8, 28, 29]), it is easy to obtain the following expressions for describing the crossovers from the 3D Mott $T^{-1/4}$ -VRH to the general soft-gap $T^{-\nu}$ -VRH with ν ranging from $1/4$ to 1 :

$$(\mathcal{F}_n(x))^{-4/3} (d\mathcal{F}_n(x)/dx) = (9\pi\beta_M E_{sg}/2k_B T_M)^{1/3} (E_{sg}/k_B T) \quad (7)$$

$$2r/\xi = [6^{1/3}/(\pi\beta_M E_{sg}/k_B T_M)^{1/3}] (\mathcal{F}_n(x))^{-1/3} \quad (8)$$

$$\eta = 2r/\xi + (E_{sg}/k_B T)x \quad (9)$$

where $x = \epsilon/E_{sg}$ with ϵ being the optimum hopping energy, and where

$$\mathcal{F}_n(x) = \int_0^x \frac{t^n dt}{1+t^n}. \quad (10)$$

For a given value of n , by solving equations (7)–(10) we will obtain the exponent η of the resistivity as a function of the temperature T . Recall that $R(T) = R_0 \exp \eta(T)$. For the particular case of $n = 2$, these equations reduce to the corresponding expressions for the 3D Mott–ES crossover of reference [8].

In the high-energy limit, $|E| \gg E_{sg}$, the DOS of equation (6) leads to $G(E) = \alpha E_{sg}^n \equiv G_0 = \text{constant}$. This is the Mott case, where $\mathcal{F}_n(x) = x$, and the solution of equations (7)–(9) gives the optimum hopping energy:

$$\epsilon_M = 3^{-3/4} (\pi G_0 \xi^3 / 6)^{-1/4} (k_B T)^{3/4} \quad (11)$$

and the Mott law of equation (1) for $R(T)$ with $\beta_M = (6/\pi)(3^{1/4} + 3^{-3/4})^4 \approx 18.1$.

In the opposite limit, $|E| \ll E_{sg}$, the DOS of equation (6) leads to $G(E) = \alpha |E|^n$. Consequently, $\mathcal{F}_n(x) = x^{(n+1)}/(n+1)$, and the solution of equations (7)–(9) gives the optimum hopping energy:

$$\epsilon_{VRH}(n) = 3^{-3/(n+4)} (n+1)^{4/(n+4)} (\pi \alpha \xi^3)^{-1/(n+4)} (k_B T)^{-3/(n+4)} \quad (12)$$

and the VRH resistance:

$$R(T) = R_0 \exp(T_0(n)/T)^{(n+1)/(n+4)} \quad (13)$$

with the characteristic VRH temperature $T_0(n)$ given by

$$T_0(n) = [2(n+4)^{(n+4)}/(9\pi(n+1)^n)]^{1/(n+1)} [\beta_M^{-1} T_M (E_{sg}/k_B)^n]^{1/(n+1)}. \quad (14)$$

For the particular case of the parabolic DOS for $n = 2$, equation (13) takes the form of the ES law of equation (2). The temperature $T_0(n) = T_0(2) \equiv T_{ES}$ of equation (14) then gives T_{ES} with $\beta_{ES} = 4 \times 6^{1/3} \approx 7.27$.

Furthermore, if the crossover temperature $T_c(n)$ is defined as the temperature for which the optimum hopping energies of the two limit regimes are equal to each other, $\epsilon_M = \epsilon_{VRH}(n)$, then from equations (11) and (12) we have

$$T_c(n) = 3(n+1)^{16/3n} (\pi/6)^{1/3} (E_{sg}/k_B)^{4/3} (\beta_M/T_M)^{1/3}. \quad (15)$$

Note that, in writing the expressions of equation (14) for $T_0(n)$ and of equation (15) for $T_c(n)$, the relation $\alpha E_{sg}^n = G_0$ was used.

It should be mentioned that, in fitting these crossover expressions to the experimental data with a defined Mott characteristic temperature T_M deduced from the data, the temperature $T^* \equiv E_{sg}/k_B$ is the only adjustable fitting parameter used. The value for the VRH characteristic temperature $T_0(n)$ obtained from equation (14) and the value for the crossover temperature $T_c(n)$ obtained from equation (15) can then be compared with corresponding experimental values found from the low-temperature resistance data.

For the experimental data for the amorphous $\text{Ni}_x\text{Si}_{1-x}$ film, exhibiting a power $\nu = 0.72$, the power of the energy in the DOS corresponds to $n \approx 6$ according to equation (5). In this case the function $\mathcal{F}_n(x)$ of equation (10) has the form

$$\mathcal{F}_6(x) = x + \frac{1}{4\sqrt{3}} \ln \frac{x^2 - \sqrt{3}x + 1}{x^2 + \sqrt{3}x + 1} - [2 \tan^{-1} x + \tan^{-1}(2x + \sqrt{3}) + \tan^{-1}(2x - \sqrt{3})]/6. \quad (16)$$

The forms of the function $\mathcal{F}_n(x)$ for the other most commonly used values of n are given in the appendix.

3D Mott to VRH Resistance Theory

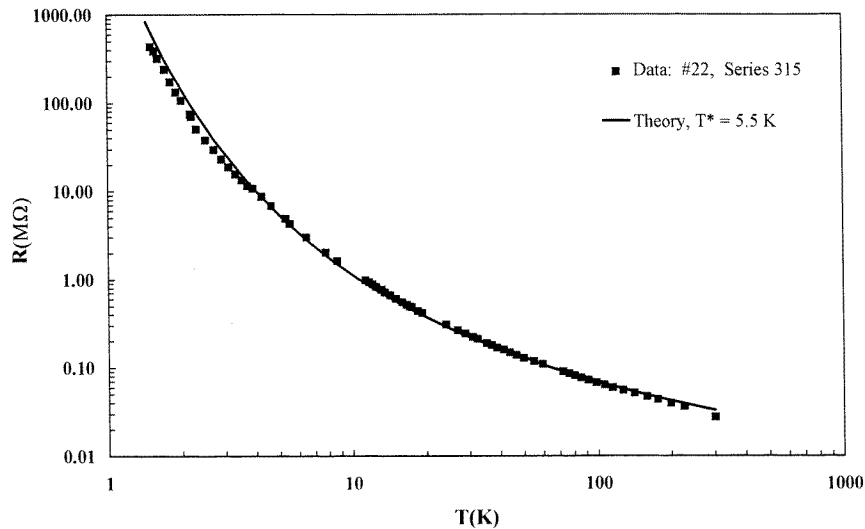


Figure 2. Comparison of the present crossover expressions with the data of figure 1. Only one fitting parameter, $T^* = E_{sg}/k_B$, is used. The curve for $T^* = 5.5$ K fits the experimental points well.

Using the experimental value of the Mott characteristic temperature $T_M = 7310$ K and $\beta_M = 18.1$ defined above, and treating $T^* = E_{sg}/k_B$ as a fitting parameter, we solve equations (7)–(9), (16) numerically. The prefactor R_0 was set to $R_0 = 0.00353 M\Omega$. The curve obtained using $T^* = 5.5$ K gives an acceptable fit to the experimental data, as seen in figure 2. Moreover, using this value of T^* , the theoretical expressions of equation (14) and of equation (15) predict for the characteristic temperature $T_0(6)$ and the crossover

temperature $T_c(6)$ the values $T_0(6) = 35.2$ K and $T_c(6) = 17.9$ K. These theoretical values should be respectively compared to experimental values of $25.2 \text{ K} \pm 2 \text{ K}$ for the characteristic temperature and of $9 \text{ K} \pm 3 \text{ K}$ for the crossover temperature deduced directly from the data. In fact, to compare with the present resistance data of $\nu = 0.72 \pm 0.03$, following the relation given as equation (5), there are two possible approximate (integer) values for the power n : $n = 6$ and $n = 7$. The lower value, $n = 6$, was chosen here for simplicity. We believe that using such an approximation for determining n will unavoidably affect the agreement between the theory and experiment, especially at low temperatures, as can be seen in figure 2.

In conclusion, we report experimental data for the smooth crossover from the 3D Mott $T^{-1/4}$ -VRH to the soft-gap $T^{-\nu}$ -VRH with $\nu \approx 0.72$, measured for an amorphous $\text{Ni}_x\text{Si}_{1-x}$ film. The data are quite well described by the simple theory proposed for general crossovers from the 3D Mott $T^{-1/4}$ -VRH to the soft-gap $T^{-\nu}$ -VRH for any ν from $1/4$ to 1 . The theory is based on the proposed DOS with a soft gap that is harder than the parabolic gap in the ES single-particle approximation. The significant role of many-particle correlations in transport processes at very low temperatures is well recognized. However, in spite of recent efforts [30, 31], this very complicated problem is still far from being understood, and currently there is no an adequate many-particle theory for VRH at very low temperatures.

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Appendix

The forms of function $\mathcal{F}_n(x)$ for some important cases are given below.

(i) For the case of $n = 3$, corresponding to $\nu \approx 0.57$:

$$\mathcal{F}_3(x) = x - \frac{1}{3} \ln \frac{1+x}{\sqrt{1-x+x^2}} - \frac{1}{\sqrt{3}} \tan^{-1} \frac{x\sqrt{3}}{2-x}.$$

(ii) For the case of $n = 4$, corresponding to $\nu \approx 0.63$:

$$\mathcal{F}_4(x) = x - \frac{1}{4\sqrt{2}} \ln \frac{1+x\sqrt{2}+x^2}{1-x\sqrt{2}+x^2} - \frac{1}{2\sqrt{2}} \tan^{-1} \frac{x\sqrt{2}}{1-x^2}.$$

(iii) For the case of $n = 5$, corresponding to $\nu \approx 0.68$:

$$\begin{aligned} \mathcal{F}_5(x) = & x - \frac{1}{5} \ln(1+x) + \frac{1}{5} \left[\cos(\pi/5) \ln(x^2 - 2x \cos(\pi/5) + 1) \right. \\ & + \cos(3\pi/5) \ln(x^2 - 2x \cos(3\pi/5) + 1) \\ & - 2 \sin(\pi/5) \tan^{-1} \frac{x \sin(\pi/5)}{1 - x \cos(\pi/5)} \\ & \left. - 2 \sin(3\pi/5) \tan^{-1} \frac{x \sin(3\pi/5)}{1 - x \cos(3\pi/5)} \right]. \end{aligned}$$

For a given form of $\mathcal{F}_n(x)$, equations (7)–(9) could easily be solved numerically by, for example, the standard Newton–Raphson method.

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