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A useful Mott–Efros–Shklovskii resistivity crossover formulation for three-dimensional films

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Abstract. Useful and simple 3D crossover expressions are presented for the resistance versus temperature behaviour in highly insulating 3D films. At high temperatures, this theory extrapolates to the Mott variable-range hopping law, and at low temperatures to the Efros–Shklovskii variable-range hopping law. Good agreement is found between the crossover theory and resistance measurements.

1. Introduction

Variable-range hopping (VRH) is a very general conduction mechanism in Anderson localized systems at sufficiently low temperatures. An introduction to VRH can be found in reference [1]. As was originally pointed out by Mott [2, 3], the VRH resistivity $\rho(T)$ can be obtained by optimizing (minimizing) the exponent

$$\eta = 2r/\xi + \epsilon/k_B T \tag{1}$$

in the hopping probability $p \propto \exp(-\eta)$, where ξ is the localization length, and where the hopping distance r and the hopping energy ϵ are related to each other through the one-particle density of states (DOS) $g(\varepsilon)$ by the 3D normalization expression:

$$\frac{4}{3}\pi r^3 \int_0^{\epsilon} g(\varepsilon)\,\mathrm{d}\varepsilon = 1 \tag{2}$$

where ε is the one-particle energy measured from E_F.

Assuming that the DOS is constant near the Fermi level E_F, and is equal to $g(E_F) = g_0$, such an optimization derivation leads to the famous Mott $T^{-1/4}$-law for the resistivity $\rho(T)$ [2, 3]:

$$\rho(T) = \rho_0 \exp(T_M/T)^{1/4} \tag{3}$$

with

$$T_M = \beta_M/(k_B \xi^3 g_0). \tag{4}$$

The temperature dependence of the pre-exponential factor ρ_0 is usually weak in comparison with the exponential term [4], and we ignore its temperature contribution. The Mott law, equation (3), has been more rigorously confirmed by Ambegaokar *et al* [5] and by Pollak [6], and is widely accepted. But there exists a considerable discrepancy as regards the values of the coefficient β_M: 1.5 given by Mott in [1], $24/\pi = 7.6$ given by Mott in [7], 18.1 given by Castner in [8], 21.1 given by Skal and Shklovskii in [9] and 27 given by Ortuno and Pollak in [10].

Pollak was the first to suggest that the long-range nature of the Coulomb interactions should lead to a dip (soft gap) in the DOS at the Fermi energy [11]. Efros and Shklovskii (ES) showed that the DOS vanishes at E_F for $T = 0$, and has the following parabolic form in the immediate vicinity of E_F [12]:

$$g(\varepsilon) = \alpha_{3D}\varepsilon^2 \tag{5}$$

with

$$\alpha_{3D} = (3/\pi)(\kappa^3/e^6) \tag{6}$$

where ε is the one-particle energy measured from E_F, $\kappa = \varepsilon_r\varepsilon_0$ is the dielectric constant (ε_0 is the permittivity constant, $=8.85 \times 10^{-12}$ C^2 J^{-1} m^{-1}, and ε_r is the relative dielectric constant), and e is the elementary charge. This DOS has been supported by computer studies [13], and has been observed in electron tunnelling experiments by Massey and Lee [14].

ES have also argued that the most observable manifestation of the Coulomb gap, expressed by equation (5), should be the temperature dependence of the VRH resistivity $\rho(T)$, which has the form [12]:

$$\rho(T) = \rho_0 \exp(T_{ES}/T)^{1/2} \tag{7}$$

with

$$T_{ES} = e^2\beta_{ES}/(k_B\kappa\xi) \tag{8}$$

where the coefficient β_{ES} is equal to 2.8 according to Shklovskii and Efros [15] or to 0.57 according to Adkins [16]. Thus, as for the case of β_M, there exists a discrepancy as regards the value of β_{ES}.

Both the Mott and ES VRH laws have been observed for different materials [8, 15]. Note that while the ES law is mostly observed for doped semiconductors and/or in magnetic fields, in principle one would expect to observe both of the laws for the same sample in different ranges of temperature, with the effective 'crossover' temperature T_c determined when the energy ε_c (the half-width of the Coulomb gap) of the parabolic DOS of equation (5) is equal to g_0:

$$T_c = \varepsilon_c/k_B \approx [(\beta_M\pi T_{ES}^3)/(\beta_{ES}^3\, 3T_M)]^{1/2}. \tag{9}$$

Massey and Lee observed a width ε_c of 0.75 meV ($T_c = 8.7$ K) for boron-doped silicon [14]. At $T > T_c$, the optimum hopping energy ϵ_{opt} is much greater than ε_c; i.e. the electrons participating in the hopping events probe only the DOS far away from the Fermi level where the DOS is relatively constant in this case. Thus, these electrons are not 'aware' of any anomalous depression of the DOS around E_F, and Mott hopping is anticipated. At $T < T_c$ where $\epsilon_{opt} < \varepsilon_c$, the electrons sample the depressed DOS, and ES hopping should prevail. Thus, a 'Mott–ES crossover' is expected; but experimentally it is difficult to distinguish between the Mott $T^{-1/4}$-law at high temperatures and the ES $T^{-1/2}$-law at low temperatures unless good quality data over several orders of magnitude of temperature are available. In fact, some ten years ago the Mott–ES crossovers had been observed experimentally for a few materials [17, 18]. Recently, the topic has again been actively studied, stimulated by a 3D theoretical crossover formalism introduced by Aharony *et al* [19]. The aim of the present work is to present useful and very simple expressions for $\rho(T)$, which describe well the Mott–ES crossover in highly insulating 3D films.

2. The theoretical crossover formalism

Theoretically, the Mott–ES crossover for 3D VRH was first studied by Aharony *et al* [19]. Recently Yigal Meir has also proposed a percolation picture for describing the 3D problem [20].

We will now derive very useful and simple 3D crossover expressions based upon the argument proposed by one of the authors for 2D systems [21]. The 3D DOS can be approximated by

$$g(\varepsilon) = \alpha_{3D}\varepsilon_c^2\varepsilon^2/(\varepsilon_c^2 + \varepsilon^2) \tag{10}$$

where $\alpha_{3D} = (3/\pi)(\kappa^3/e^6)$ [12, 15]. In the limit of large ε, the DOS approaches the constant value of g_0; and in the opposite limit of small ε, the DOS approaches a parabolic dependence. Efros was perhaps the first to suggest the approximation of the DOS given by equation (10) [22]. Note that equation (10) does not take into account the 'weakening' or 'smearing' of the gap due to finite-temperature effects. If the temperature is raised above zero, 'thermal smearing' of the Fermi surface will raise the DOS in the gap from zero at E_F to a finite value, which is stated by Mott and Kaveh [23] to be of the order of $k_BT/\varepsilon_{bw}^2a^3$; ε_{bw} is the bandwidth and a is the distance between neighbouring impurity sites. At sufficiently high temperatures, the Coulomb gap will be 'closed' or absent, and the Mott $T^{-1/4}$-law will apply. One experimental study has recently been reported on this subject [24]. We have neglected this second-order correction in order to simplify the mathematics. Van Keuls *et al* have examined the transition from the ES hopping regime to the screened Mott hopping regime [25].

Using the suggested 3D DOS expression of equation (10), we find the following 3D crossover expressions by optimizing (minimizing) the probability exponent η (using $\mathrm{d}\eta/\mathrm{d}r = 0$ or $\mathrm{d}\eta/\mathrm{d}\epsilon = 0$):

$$(\epsilon^2/\varepsilon_c^2)(1 + \epsilon^2/\varepsilon_c^2)^{-1}(\epsilon/\varepsilon_c - \tan^{-1}(\epsilon/\varepsilon_c))^{-4/3} = 2^{-1/3}\,3\beta_{ES}(T_c^2/T_{ES}T) \tag{11}$$

$$2r/\xi = (6/\pi\beta_M)^{1/3}(T_M/T_c)^{1/3}(\epsilon/\varepsilon_c - \tan^{-1}(\epsilon/\varepsilon_c))^{-1/3} \tag{12}$$

$$\eta = 2r/\xi + (\epsilon/\varepsilon_c)T_c/T \tag{13}$$

and

$$R(T) = R_0\exp(\eta) \qquad \text{or} \qquad \rho(T) = \rho_0\exp(\eta). \tag{14}$$

R_0 (or ρ_0) is a constant used in the fitting to the resistance (resistivity) data. T_c is defined by equation (9). Note that in the calculations, the inverse tangent function $\tan^{-1}x$ defines an angle that must be expressed in radians and not in degrees. One first calculates a value for T_c (or ε_c) using equation (9) with values of T_M and T_{ES} extracted from the data. Then, guessing a value for ϵ/ε_c that typically ranges from 1 to 100, one calculates an experimental measurement temperature T using equation (11). Using the same value of ϵ/ε_c, one calculates the $2r/\xi$ contribution via equation (12). Next, one evaluates the argument η via equation (13) using the results of equations (9), (11), and (12). Lastly, one calculates the resistance versus the temperature using equation (14). There are three free fitting parameters, β_{ES}, β_M, and R_0. T_M and T_{ES} are extracted from the resistance data at high and at low temperatures.

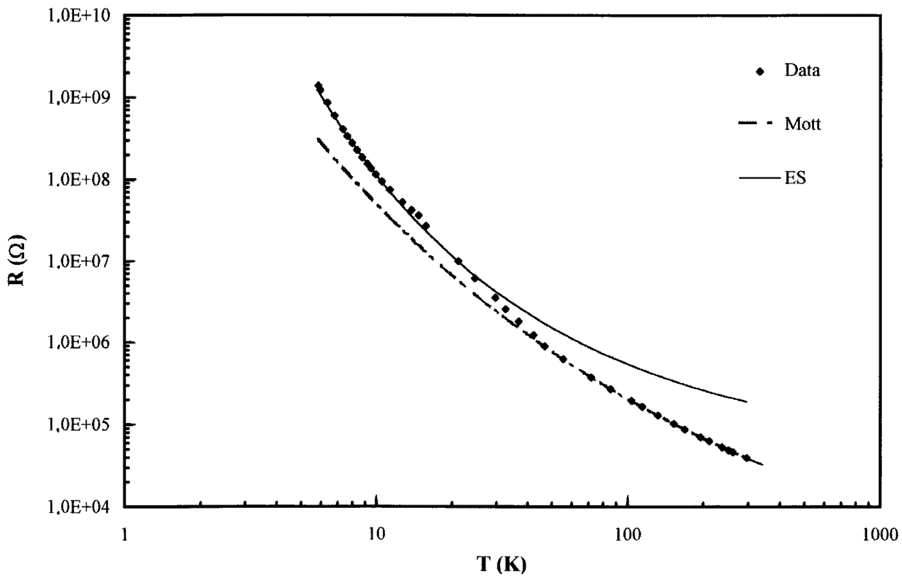

Figure 1. Resistance versus temperature for a 3D highly insulating a:Ni_xSi_{1-x} film. The dashed and solid lines represent fits to the data given by the Mott and ES VHR hopping laws. The crossover region lies between 30 K to 40 K.

In the high-temperature limit where the hopping energy ϵ approaches large values and the DOS $g(\varepsilon)$ takes on the constant value of g_0, the above theory predicts the following Mott expressions:

$$r_{opt}(T)/\xi = [3^{1/2}/(2^{3/4}\pi^{1/4}\beta_M^{1/4})](T_M/T)^{1/4} \quad (15a)$$

$$\epsilon_{opt} = [2^{1/4}/(3^{1/2}\pi^{1/4}\beta_M^{1/4})]k_B T(T_M/T)^{1/4} \quad (15b)$$

and

$$\eta = [3^{1/2} + 3^{-1/2}][2/(\pi\beta_M)]^{1/4}[T_M/T]^{1/4} \quad (15c)$$

with $T_M = \beta_M/(k_B g_0 \xi^3)$.

If one chooses $\beta_M = 18.1$, then the above three equations simplify to

$$r_{opt}(T)/\xi \approx 0.375(T_M/T)^{1/4} \quad (16a)$$

$$\epsilon_{opt} \approx 0.25 k_B T(T_M/T)^{1/4} \quad (16b)$$

and

$$\eta \approx [T_M/T]^{1/4} \quad (16c)$$

which yields directly the Mott $T^{-1/4}$ law for VRH. For the Mott law to be valid, $r_{opt}(T)/\xi > 1$.

In the low-temperature limit of small hopping energies ϵ, the theory predicts the ES expressions:

$$r_{opt}/\xi = [3^{1/6}/(2^{5/6}\beta_{ES}^{1/2})](T_{ES}/T)^{1/2} \quad (17a)$$

$$\epsilon_{opt} = [6^{1/6}/\beta_{ES}^{1/2}]k_B T(T_{ES}/T)^{1/2} \quad (17b)$$

and

$$\eta = [6^{1/6}2/\beta_{ES}^{1/2}][T_{ES}/T]^{1/2} \quad (17c)$$

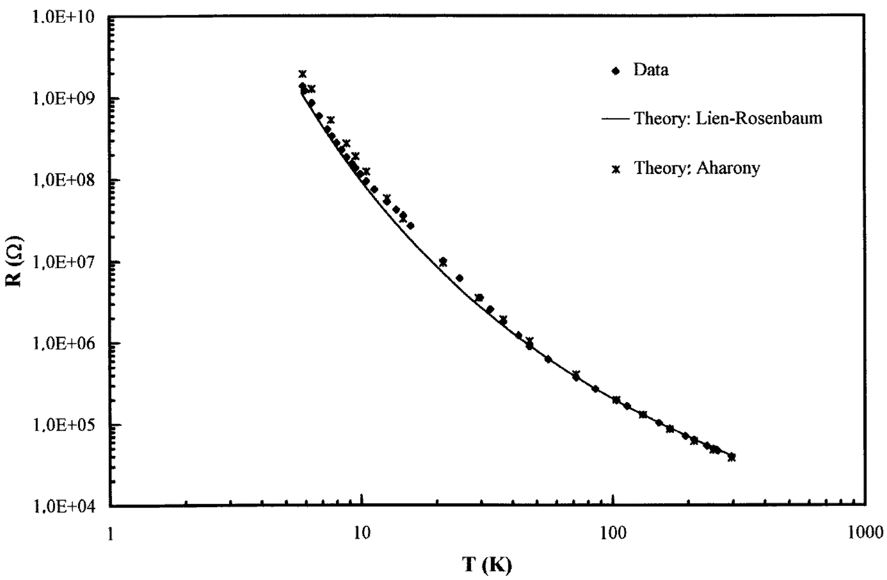

Figure 2. Comparison of our theory and also of the Aharony theory to the data on the a:Ni_xSi_{1-x} film. The same two fitting parameters, the Mott temperature T_M and the Efros–Shklovskii temperature T_{ES}, were used in each theory. T_M and T_{ES} were extracted from the data of figure 1.

with $T_{ES} = \beta_{ES}e^2/(k_B\kappa\xi)$.

If one chooses $\beta_{ES} = 7.27$, then the above three expressions simplify to

$$r_{opt}/\xi \approx 0.25(T_{ES}/T)^{1/2} \tag{18a}$$

$$\epsilon_{opt} \approx 0.5k_BT(T_{ES}/T)^{1/2} \tag{18b}$$

and

$$\eta \approx [T_{ES}/T]^{1/2} \tag{18c}$$

which yields directly the ES $T^{-1/2}$-law for VRH. For the ES law to be valid, $r_{opt}/\xi > 1$.

Our results differ from the 3D expressions derived by Aharony *et al* [19]. The Aharony argument consists of writing the hopping energy ϵ as the sum of two energies: $\epsilon = t_1/g_0r^3 + t_2e^2/\kappa r$, where t_1 and t_2 are constants, and then minimizing η with respect to r, which results in an explicit functional dependence of $\ln R$ on T [19]. We believe that our DOS approach describes the physics more accurately.

3. The data analysis procedure, film fabrication, data, and comparison with the theories

The exponent y and the effective temperature T_0 in the general VRH resistance expression $R(T) = R_0\exp[(T_0/T)^y]$ can be simply determined from the data using a technique described by Hill [26], and later by Zabrodskii and Zinov'eva [27]. If one calculates values of $w(T) = -\mathrm{d}\ln R/\mathrm{d}\ln T = y(T_0/T)^y$ from the resistance data $R(T)$, and then makes a linear regression fit through the $\log(w)$ versus $\log(T)$ data, the slope of the linear regression line fit is equal to the exponent y; and the intercept of the line fit, I, is related to the effective temperature T_0 via the expression $T_0 = (10^I/y)^{1/y}$. Thus, one can readily

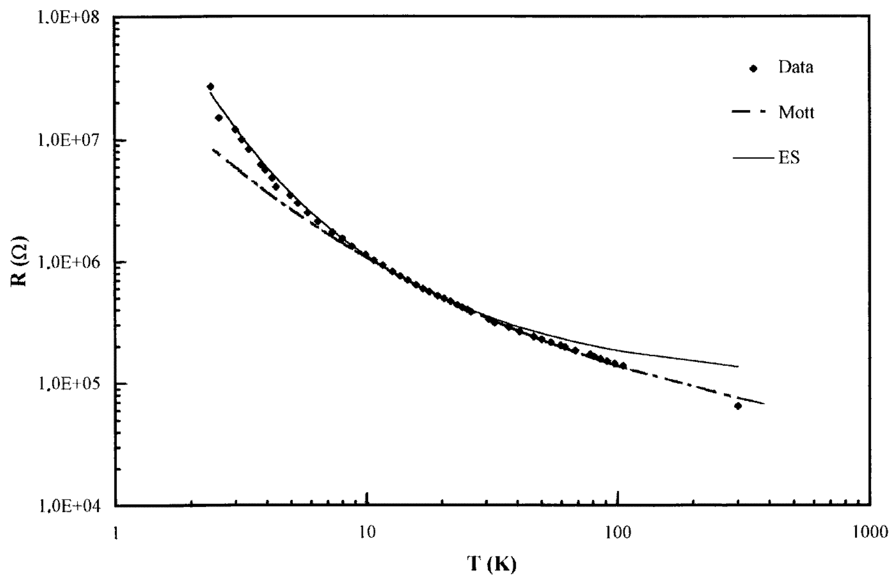

Figure 3. Resistance versus temperature for a 3D moderately insulating a:In_xO_y film, fabricated using thermal evaporation of indium oxide powder. The dashed and solid lines represent fits to the data given by the Mott and ES VHR hopping laws. The crossover region lies between 10 and 30 K.

determine whether the resistance data display the Mott $T^{-1/4}$-law or the ES $T^{-1/2}$-law, or another VRH dependence.

Insulating films were prepared by evaporating appropriate materials onto glass substrates held at room temperature. Figure 1 shows the resistance of an amorphous a:Ni_xSi_{1-x} film versus temperature. This film comes from a series fabricated by co-evaporating Ni and Si using two electron guns. EDAX (energy-dispersive analysis of x-rays) yielded a nickel content $x \approx 8$ at.% Ni. The metal–insulator transition for this system is located at $x_c \approx 24$ at.% Ni; thus, this film is located in the deeply insulating region. The geometric factor f_g needed to convert the resistance R to resistivity ρ is $f_g = 1.99 \times 10^{-6}$ cm for this film which is 1040 Å thick; the conversion to ρ is not necessary for calculating the w-values. The 'w'-technique yielded a Mott-type VRH law $R(T) = 236 \exp[(171\,060/T)^{0.26}]$ in Ω for temperatures between 300 K to 42 K, and for temperatures below 25 K an ES-type VRH law going as $R(T) = 42\,700 \exp[(671/T)^{0.49}]$ in Ω. These two expressions are compared to the data in figure 1. Using $T_M = 171\,060$ K, $T_{ES} = 671$ K, $\beta_M = 18.1$, and $\beta_{ES} = 7.27$, this theory and the Aharony theory [19] are compared to the data as shown in figure 2. Both crossover theories give good fits to the resistance data on this highly insulating film. The theoretical crossover temperature T_c is 9.3 K, similar to the value observed for boron-doped silicon [14]. Note that the criterion $r_{opt}/\xi \geqslant 1$ is satisfied in both the Mott and ES temperature regimes.

Data from an amorphous In_xO_y film were also analysed. This film was prepared by thermal evaporation of In_xO_y in a partial atmosphere of O_2 of 6×10^{-5} mm Hg [18]. This film has a thickness of 460 Å and a geometry factor $f_g = 5.11 \times 10^{-6}$ cm. The 'w'-technique yielded Mott and ES exponents of 0.27 and 0.57; since these exponents were sufficiently close to the theoretical exponents 1/4 and 1/2, a Mott $T^{-1/4}$-law and an ES $T^{-1/2}$-law were forced through these data, as shown in figure 3. For this case,

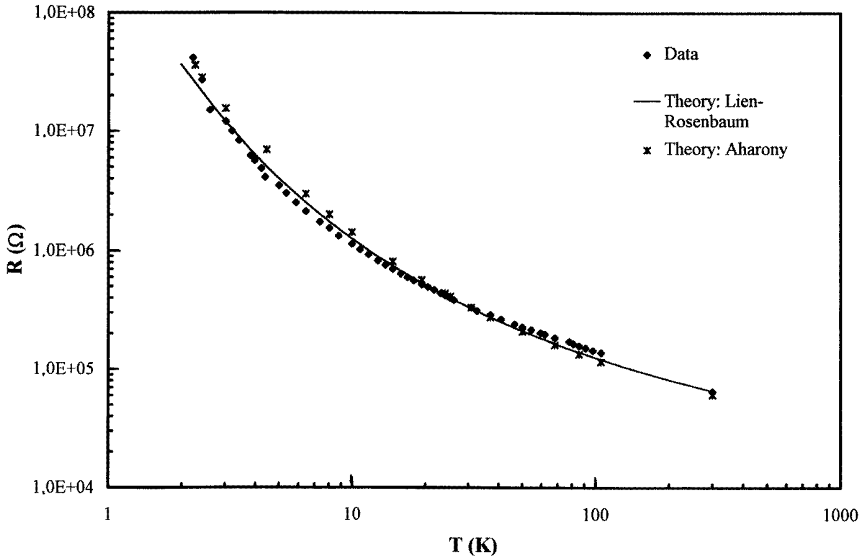

Figure 4. Comparison of our theory and also of the Aharony theory to the data on the a:In_xO_y film of figure 3; our theory gives a better fit to the data than does the Aharony theory.

$T_M = 4400$ K and $T_{ES} = 78.2$ K, and the resulting theoretical crossover temperature T_c is 2.3 K. This film is much less insulating when compared to the amorphous NiSi film; and the criterion $r_{opt}/\xi \geqslant 1$ is satisfied only in the lower-temperature regimes of the Mott and of the ES fitting intervals. Yet, this crossover theory using $\beta_M = 18.1$ and $\beta_{ES} = 7.27$ yields a fairly good fit to the In_xO_y data, as seen in figure 4.

Data from a second indium oxide film were compared to the theory. This amorphous film was prepared using a completely different method, in which an indium target was sputtered using an argon beam in the presence of a background pressure of oxygen [28]. This film has a thickness of 1120 Å and a geometric factor f_g of 1.61×10^{-6} cm. The resistance versus temperature data were collected in two different runs, using a 'continuous-flow' cryostat for data taken above 5 K, and an immersion cryostat below 4.2 K. There is a discontinuity in the 4.2 K resistance values between the two runs owing to an 'aging' effect with room temperature cycling. The resistance is sensitive to the oxygen content, since oxygen easily diffuses either into or out of the indium oxide film at room temperature, making the film more resistive or conductive [29]. Between 300 K and 30 K, a Mott VRH law $R_M(T) = 17\,700 \exp[(16\,630/T)^{0.245}]$ in Ω gave an excellent fit to the data; and between 40 K and 5 K, an ES VRH law $R_{ES}(T) = 227\,000 \exp[(116/T)^{0.56}]$ in Ω gave a good fit, as illustrated in figure 5. Using the above temperatures T_M and T_{ES}, the theory gives a nice fit between 5 K and 300 K as shown in figure 6; and the discontinuity between the two data runs is clearly evident below 4.2 K, where the crossover formula is used to predict extrapolated resistance values below 4.2 K. The theoretical crossover temperature of 2.2 K is again too low, compared to the experimental one of about 35 K.

Since the values of $\beta_M = 18.1$ and $\beta_{ES} = 7.27$ gave very acceptable fits to the three R versus T data sets, we have not attempted to vary these parameters in order to improve and optimize the fits. It is interesting to note that the more insulating the films, the better the agreement of the two theories with the data.

In conclusion, our theory works as well as or better than the Aharony crossover

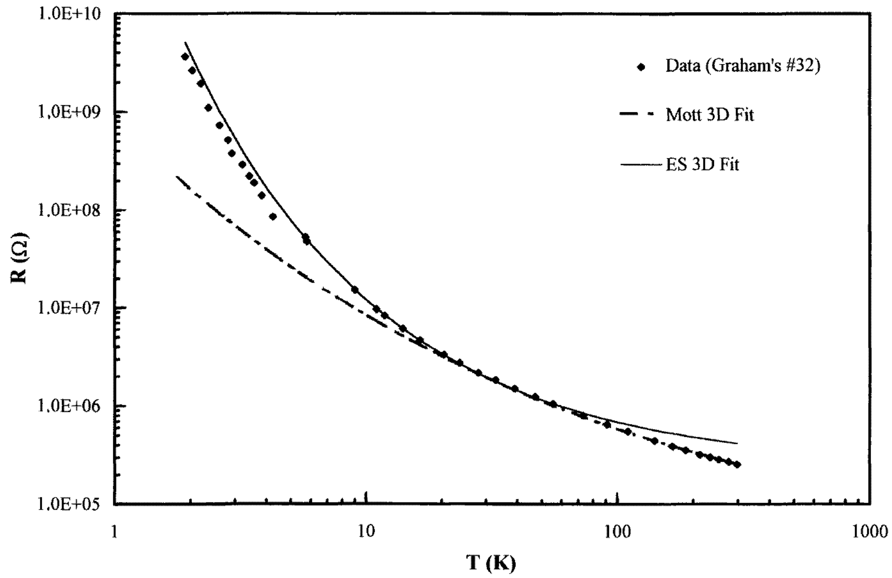

Figure 5. Resistance versus temperature for a 3D strongly insulating a:In_xO_y film, fabricated by sputtering an indium target. The dashed and solid lines represent fits to the data given by the Mott and ES VHR hopping laws. The crossover region lies between 20 K and 40 K.

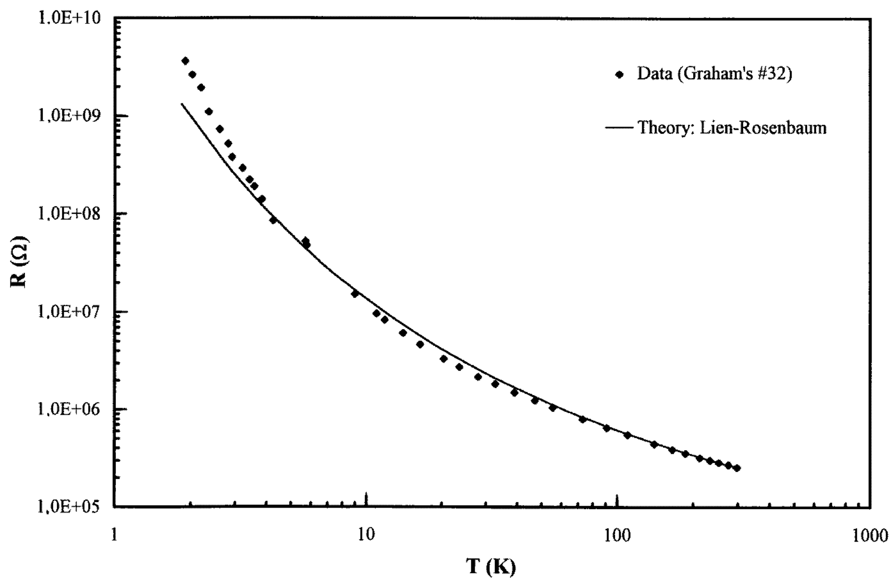

Figure 6. Comparison of our theory to the data on the a:In_xO_y film of figure 5. This theory gives an acceptable fit to the data; the discontinuity below 4 K arises from the 'cycling and aging' effects of different runs, owing to diffusion of oxygen into or out of the film at room temperature.

expression [19]. Its success lies in the reasonable approximation for the single-particle density of states given by equation (10), and in using the condition that the localization length ξ must be the same in the two regimes. However, there remain two problems. The theoretical values for the crossover temperatures T_c are a factor of three to fifteen times smaller than the values observed experimentally. An equally serious discrepancy is the estimation for the localization length ξ using the expression $T_{ES} = \beta_{ES} e^2/(k_B \kappa \xi)$; if we use a value of 14 for the relative dielectric constant [30], ξ takes on the value of 1600 Å for the amorphous nickel silicon film, and about 14 000 Å for the amorphous indium oxide films. These values should be compared to values of 50 Å to 100 Å estimated for indium oxide by Ovadyahu [29]. This discrepancy possibly casts doubts on the validity of the ES model. Other physical processes could lead to a parabolic DOS around E_F. However, the one fitting parameter that directly depends upon the ES model is the crossover temperature T_c through its dependence upon T_{ES}. On one hand, the crossover expressions all involve T_c and T_{ES}; and these expressions yield good agreement with the experimental data. On the other hand, the expression for T_{ES} predicts unreasonable magnitudes for the localization length. It is not clear what the reason for this discrepancy is. Similar discrepancies in the magnitude of $\kappa\xi$ have been reported by Zhang *et al* [31, 32] and by Ionov *et al* [33]. Adkins has also pointed out inconsistencies, using the ES model to describe VRH phenomena in granular materials and 3D composites [16, 34].

In addition, a magnetic moment of nickel could lead to a 'hard' gap in the DOS, resulting in simple exponential activation at low temperatures [35, 36]. We see no evidence for a crossover to a '$y = 1$' hopping law. Moreover, there is recent evidence that very small clusters of Ni atoms are non-magnetic. From magnetoconductance data taken on thin copper films deposited upon very small isolated Ni spheres of controlled diameter, Lin *et al* concluded that Ni spheres, having diameters smaller than 8 Å, possess no magnetic moment [37]. Beckmann and Bergmann also observed that pairs of Ni atoms have a magnetic moment, while single Ni atoms are non-magnetic [38]. In view of the low nickel content in our a:Ni_xSi_{1-x} film, there should be no magnetic moments and no complications arising from them.

We do not know what the role of impurity bands and Hubbard bands is in determining the DOS in these amorphous materials. The optical joint density of states (OJDOS) observed by Bayliss *et al* [30] does not appear to be relevant to this problem, since their observed gap of about 1 eV (12 000 K) would lead to simple activation even at room temperature.

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