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Experimental study of the Ioffe–Regel criterion for amorphous indium oxide films

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Abstract. The Ioffe–Regel criterion predicts the existence of a metal–insulator transition in a film series when the parameter $k_F l_e$ satisfies the criterion $k_F l_e \approx 1$; here k_F is the Fermi wavenumber and l_e is the elastic mean free path of the carriers. According to this criterion, films having $k_F l_e \gg 1$ are metallic, while films having $k_F l_e \ll 1$ are insulating. We experimentally observe the metal–insulator transition in amorphous indium oxide films at $k_F l_e = 5.2 \pm 0.5$. The values of $k_F l_e$ were calculated from room temperature resistivity and Hall voltage measurements, while the metal–insulator transition was determined from low-temperature resistivity data using the 'w'-criterion of Mobius and of Zabrodskii and Zinov'eva.

1. Introduction

Ioffe and Regel predicted that there should be a metal–insulator transition when the criterion $k_F l_e \approx 1$ is satisfied [1]. Here $k_F = (3\pi^2 n)^{1/3}$ is the Fermi wavenumber expressed in terms of the carrier density n, and $l_e = v_F \tau_e = \hbar k_F \tau_e / m^*$ is the elastic mean free path of the free carriers. Using the Boltzmann conductivity expression $\sigma = ne^2\tau_e/m^*$ to eliminate τ_e/m^*, one obtains for the parameter $k_F l_e$:

$$k_F l_e = \hbar(3\pi^2)^{2/3}/(e^2 \rho n^{1/3}) \tag{1}$$

where ρ is the resistivity. The carrier concentration n can be estimated from Hall voltage measurements:

$$V_H = IB/ent. \tag{2}$$

Here I is the current through the film, B is the magnetic field applied perpendicular to the film, and t is the film thickness.

Equation (1) is valid only for weakly disordered metallic films where the Fermi energy level E_F and Fermi wavenumber k_F are well defined. As the metal–insulator transition is approached, these criteria may no longer be fulfilled, although we continue to use equation (1) with caution for lack of a better formalism. The use of the Boltzmann expression in the derivation of equation (1) takes no account of the low-temperature quantum corrections to the conductivity from weak-localization (WL) and electron–electron interaction (EEI) effects. The weak-localization effects can cause a significant reduction in the conductivity from its room temperature value. The appropriate expressions involving $\sigma, k_F l_e$, and the inelastic scattering times (or equivalently, the inelastic lengths) have been derived by Kaveh and Wiser [2] and discussed by Lee and Ramakrishnan [3]. Lee and Ramakrishnan also derive

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the conductivity corrections arising from the electron–electron interactions [3]. Similarly, corrections to the Hall constant from these effects as described by Altshuler *et al* [4, 5] are not considered here. However, in this study we are primarily interested in evaluating $k_F l_e$ close to the MIT; and for the relevant metallic and weakly insulating samples, no temperature dependence of the Hall constant is observed, and the temperature dependence of the resistivity is less than 10% over the entire temperature range. For the purposes of this work, $k_F l_e$ in equation (1) may therefore be considered as effectively temperature independent for the sample range of significance, and for convenience we evaluate it from room temperature data.

If one takes the Ioffe–Regel criterion literally, namely $k_F l_e = 1$, there is a problem of interpretation. Recalling that $k_F \approx \pi/a$ where a is a typical distance between atomic sites, the elastic length $l_e = a/\pi$ is *considerably smaller* than the typical interatomic spacing, a. But a real elastic length can never be *less* than the typical atomic spacing, since there are no atoms present at distances less than the interatomic spacing a from which the carriers can scatter elastically. Thus, a more reasonable Ioffe–Regel criterion, which we propose, might be this one:

$$k_F l_e \approx \pi \tag{3}$$

rather than $k_F l_e \approx 1$ since, in this case, $l_e \approx a$. Mott has also suggested $k_F l_e \approx \pi$ [6]. According to Elliott, the wave function of the carrier loses phase memory from atom to atom if the 'elastic mean free path' *is less* than the interatomic spacing; and the carrier becomes *localized*, resulting in a transition to the insulating state [7].

It is worth pointing out that the Mooij criterion [8] may be considered as a special case of the Ioffe–Regel condition. If one calculates a resistivity ρ using a typical metallic carrier density n and using $k_F l_e = 1$ from equation (1), then the Mooij resistivity has a typical value of 2×10^{-5} Ω m, at which the slope of the resistivity versus temperature is essentially zero. Films that have resistivities less than this value have positive slopes, and films with resistivities greater than this value have negative slopes. The slope of the low-temperature resistivity in many materials arises owing to a competition between phonon scattering and between weak-localization and electron–electron interaction effects, as demonstrated by Tsuei [9]. Tsuei showed that there is nothing universal about the Mooij resistivity value, and that the crossover resistivity can vary between 0.3×10^{-6} to 5×10^{-6} Ω m from system to system [9].

We are not aware of a critical experimental test of the Ioffe–Regel criterion. It should be possible to test this criterion using low-temperature resistivity data taken on the same series of films on which high-quality room temperature Hall voltage measurements are made. The metal–insulator transition can be determined from the low-temperature resistivity data as now explained.

2. Analysis techniques near the metal–insulator transition

Since most theories predict values for conductivities rather than for resistivities, we now refer to conductivities.

Thin films may be classified as being either insulating or metallic. Insulating films are defined as exhibiting zero conductivity at absolute zero in temperature. In contrast, metallic films always display finite conductivities at absolute zero.

Strongly insulating films exhibit an activated hopping conductivity which can be described by the variable-range hopping (VRH) expression:

$$\sigma(T) = \sigma_0 \exp(-(T_0/T)^y) \tag{4}$$

where σ_0 is the prefactor, T_0 is a characteristic temperature, and y is an exponent.

In contrast, the conductivity of a *3D metallic* film at sufficiently low temperatures can be described by a power-law expression:

$$\sigma(T) = \sigma(0) + CT^z \tag{5}$$

where $\sigma(0)$ is the finite zero-temperature conductivity, C is a prefactor, and z is the exponent of the temperature power law. Equation (5) might approximate the conductivity contributions of the 3D electron–electron interaction theory [10] and/or of the 3D weak-localization theory [11].

A useful technique for identifying the metal–insulator transition was previously introduced in references [12–14] and [15]. The mathematical function $w(T)$ exhibits distinctively different temperature behaviours for insulating and metallic films:

$$w(T) = \mathrm{d}\ln\sigma/\mathrm{d}\ln T = (T/\sigma)\mathrm{d}\sigma/\mathrm{d}T = -\mathrm{d}\ln\rho/\mathrm{d}\ln T. \tag{6}$$

For *strongly insulating* films exhibiting variable-range hopping conductivity, inserting equation (4) into equation (6) yields

$$w(T) = y(T_0/T)^y \tag{7}$$

which gives $w(T)$ *increasing to infinity* as the temperature approaches absolute zero. By making a linear regression fit of the $\ln[w(T)]$ versus $\ln T$ data, one can directly extract values for the hopping exponent y and the characteristic temperature T_0 using equation (7) [15]. The slope of the straight-line fit is equal to the exponent y, and the intercept I of the fit is related to the characteristic temperature via $T_0 = (\mathrm{e}^I/y)^{1/y}$.

For *3D metallic* films exhibiting slowly decreasing conductivities with decreasing temperatures, equation (5) can be substituted into equation (6) to yield

$$w(T) = zCT^z/[\sigma(0) + CT^z] = zCT^z/\sigma(T). \tag{8}$$

Observe that if the film is indeed *metallic* and exhibits a finite positive conductivity $\sigma(0)$ at absolute zero, then $w(T)$ *should extrapolate to zero* at absolute zero [12]. For these metallic cases, linear regression fits of the $\ln(w\sigma)$ versus $\ln T$ data yield values for the exponent z and the prefactor C using equation (8). Values for $\sigma(0)$ follow by evaluating equation (5) with the data. Extrapolation of $\sigma(0)$ to zero as a function of the disorder control parameter will yield another estimation for the critical value of the control parameter at the metal–insulator transition (MIT) [14].

Samples might also exhibit *temperature-independent* values of w. Such a temperature-independent behaviour of w can be realized only if $\sigma(0)$ is set to zero in equation (8). For this case, the film is *weakly insulating*, since $\sigma(T) \to 0$ as $T \to 0$ K [16]; and the conductivity data can be described using the simple temperature power-law expression:

$$\sigma(T) = CT^z \tag{9}$$

with C and $z = w$ being the two fitting parameters. Note that a variable-range hopping law *cannot* be fitted successfully to conductivity data that exhibit w-values which are *temperature independent*. We refer to films having conductivities described by equation (9) as 'weakly insulating' films [17].

3. Film preparation and characterization

Amorphous indium oxide films of about 120 nm thicknesses were prepared using an ion-beam sputter-deposition system [18]. A beam of argon ions was accelerated by an ion gun and then neutralized. This beam was directed onto a water-cooled high-purity indium

circular disc, which sputtered the indium at a rate of 0.1 nm s^{-1}. Oxygen, which was used in the reactive deposition of indium oxide, was introduced directly into the chamber as a background gas. The partial pressure of the oxygen varied in the range of 3.2×10^{-4} to 7×10^{-4} mbar. This material is doped by oxygen vacancies; the higher the partial pressure of the oxygen, the more resistive were the films. The indium oxide was deposited onto a silica slide through a mask that defined a standard Hall probe geometry. The silica substrate was maintained at room temperature during the evaporation to prevent crystallization to the In_2O_{3-x} phase. The geometric factor needed to convert resistance to resistivity was $f_g = 1.6 \times 10^{-8}$ m.

The films were measured in various low-temperature cryostats, including a ^{3}He refrigerator and a dilution refrigerator. Care was taken to prevent Joule heating of the films in the ^{3}He and dilution refrigerators. A compromise always had to be made between accuracy of the resistance measurements versus detrimental heating effects of the films below 1 K, which is reflected in the large scatter in the 'w' versus T and in the R versus T curves in the ^{3}He and mK regimes.

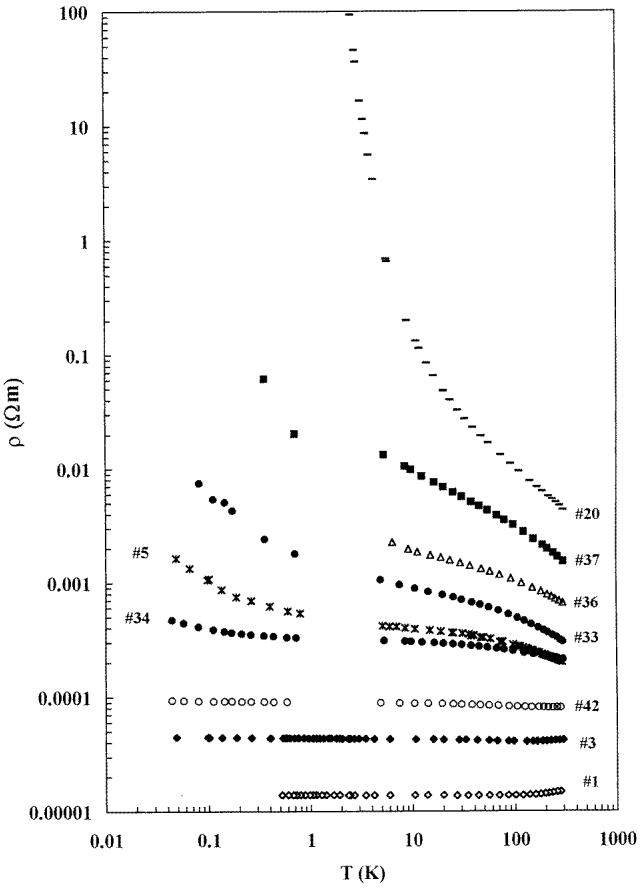

Figure 1. The resistivity versus temperature dependence for the majority of the amorphous indium oxide samples studied. For clarity, not all the films are shown. A metal–insulator transition is present in this series.

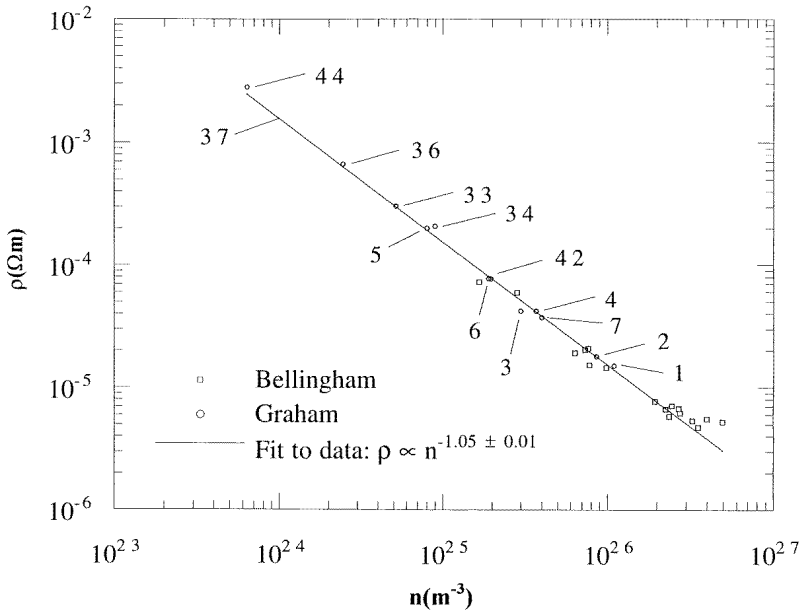

Figure 2. The room temperature resistivities versus electron carrier concentrations n of the films in the current study (labelled with the film numbers) and those of Bellingham's films.

4. Experimental results and the determinations of the metal–insulator transition and the Ioffe–Regel criterion

The resistivity versus temperature dependence of many of the indium oxide films studied is shown in figure 1. There is clearly a metal–insulator transition somewhere between the most metallic film, No 1, and the most insulating film, No 20. Figure 2 shows the room temperature resistivities of the films including those of Bellingham [18, 19] versus electron carrier concentration n, deduced from the Hall voltage measurements. The fit excludes the two points having the highest carrier concentrations. Interestingly, over the sample range studied here, the mobility scarcely changes with carrier concentration. This is a result of near cancellation of the effects of increasing the number of scattering centres (oxygen vacancies), and the consequent increasing of the wavenumber and enhanced screening. A detailed discussion is given by Bellingham *et al* [18].

Values for the parameter $k_F l_e$ calculated from equation (1) as a function of electron carrier concentration n are shown in figure 3 and are based upon room temperature Hall voltage and resistance measurements. Owing to the small electronic carrier concentration in amorphous indium oxide, the Hall voltage is relatively large and can be measured *easily* with an accuracy of ±5% [20]. Thus the parameter $k_F l_e$ is known to an accuracy of ±5%. The Hall voltage in indium oxide is unique in that it does not diverge at the metal–insulator transition (MIT), as reported by Tousson and Ovadyahu [21, 22]; this behaviour is in contrast to theoretical predictions by Wang *et al* [23] and by Bergman *et al* [24] of divergences. But the non-divergent Hall constant is consistent with a prediction by Shapiro and Abrahams [25] and makes indium oxide an ideal candidate for testing the Ioffe–Regel criterion. A good review of the diverse experimental Hall coefficient results can be found in the article by Dai *et al* [26], and a useful theoretical review is given by Friedman [27].

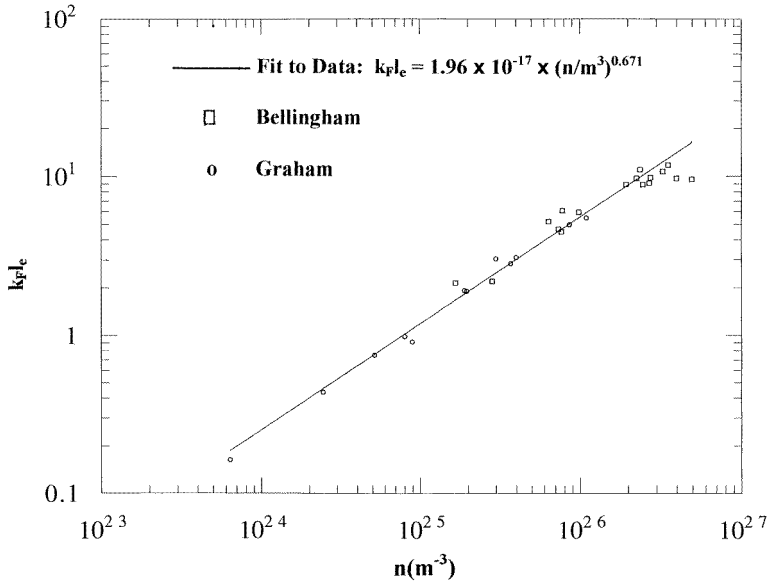

Figure 3. The parameter $k_F l_e$ versus the electron carrier concentration n.

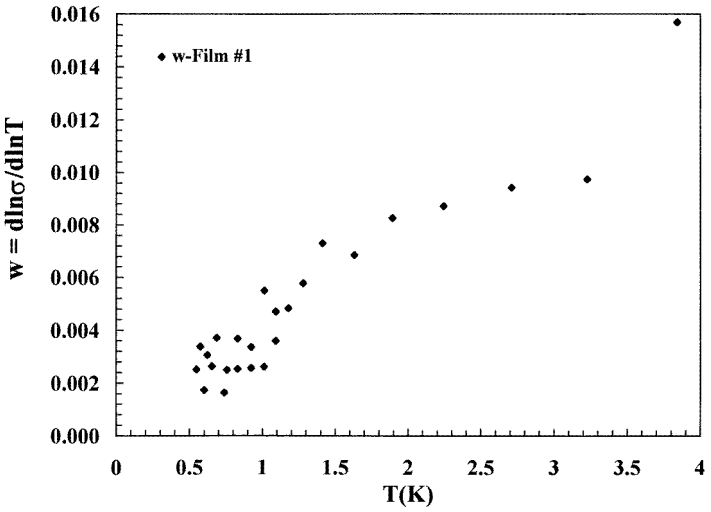

Figure 4. A plot of $w = \mathrm{d}\ln\sigma/\mathrm{d}\ln T$, the logarithmic derivative, against temperature for film No 1. w *appears* to extrapolate *to zero* below 4 K, thus identifying this film as *metallic*.

The room temperature resistivities ρ and electron carrier concentrations n are summarized in table 1.

In order to identify which films are metallic and which films are insulating, we use the criterion that the w-values must extrapolate *to zero* as $T \to 0$ for the *metallic* films [12]. In figure 4, the w-values of film No 1 *appear* to extrapolate *to zero* below 4 K, identifying this film as *metallic*. In figure 5, the conductivity data for film No 1 are compared to the metallic empirical expression $\sigma(T)/(\Omega^{-1}\ \mathrm{m}^{-1}) = 40\,760 + 226(T/\mathrm{K})^{0.83}$.

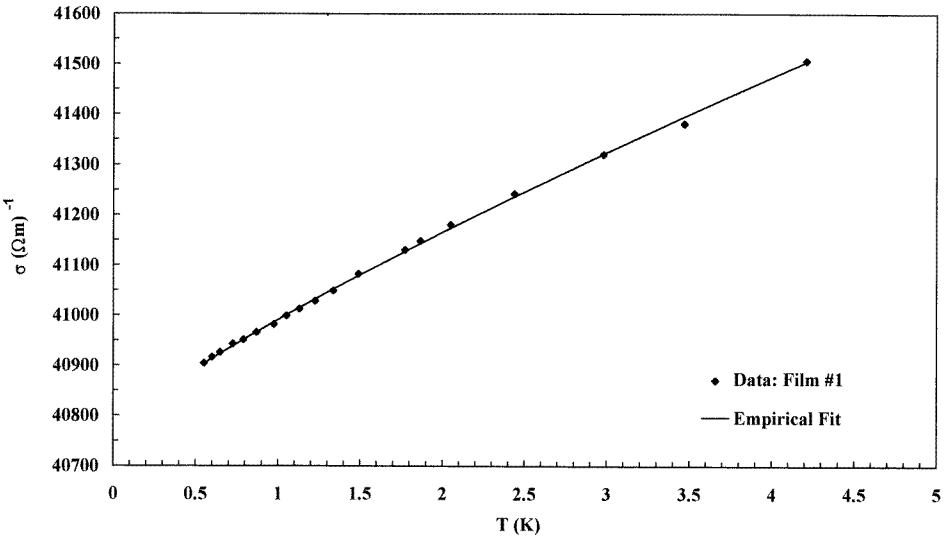

Figure 5. A fit of the metallic empirical conductivity expression $\sigma(T) = \sigma(0) + CT^z$ to the conductivity data for film No 1. See the text for fitting details.

Table 1. Film numbers, room temperature resistivities ρ, carrier concentrations n, and values for $k_F l_e$.

Film number	ρ (Ω m)	n (m^{-3})	$k_F l_e$
1	1.49×10^{-5}	1.1×10^{26}	5.5
2	1.85×10^{-5}	8.7×10^{25}	4.8
7	3.70×10^{-5}	4.0×10^{25}	3.1
4	4.05×10^{-5}	3.7×10^{25}	2.9
3	4.17×10^{-5}	3.0×10^{25}	3.0
42	7.69×10^{-5}	1.9×10^{25}	1.9
34	2.00×10^{-4}	8.0×10^{24}	0.94
5	2.08×10^{-4}	8.9×10^{24}	0.93
33	3.03×10^{-4}	5.2×10^{24}	0.76
36	6.67×10^{-4}	2.5×10^{24}	0.44

In contrast, all the remaining films, excluding film No 1, appear to be 'insulating'. Their behaviours as regards both w and σ with temperature are very similar to those observed in insulating *amorphous* nickel–silicon films located *just below* the metal–insulator transition (MIT) [16]. With reference to these amorphous Ni_xSi_{1-x} insulating films located just below the metal–insulator transition (MIT), the w-values exhibited either a *temperature-independent behaviour* over a wide temperature range or a slowly decreasing linear dependence upon temperature with a *finite intercept* at $T = 0$. Both behaviours imply insulating behaviour.

For example, as shown in figure 6, the w-values for the amorphous In_xO_y film No 2 below 4 K *decrease* but do *not* extrapolate *to zero* as $T \rightarrow 0$ K as predicted for a metallic film. Instead the w-values tend to the *finite* value of about 0.0014 as seen in figure 6. Clearly this film is insulating since the metallic criterion that $w \rightarrow 0$ as $T \rightarrow 0$ is not satisfied. If one assumes below 4 K that w *decreases linearly* with the

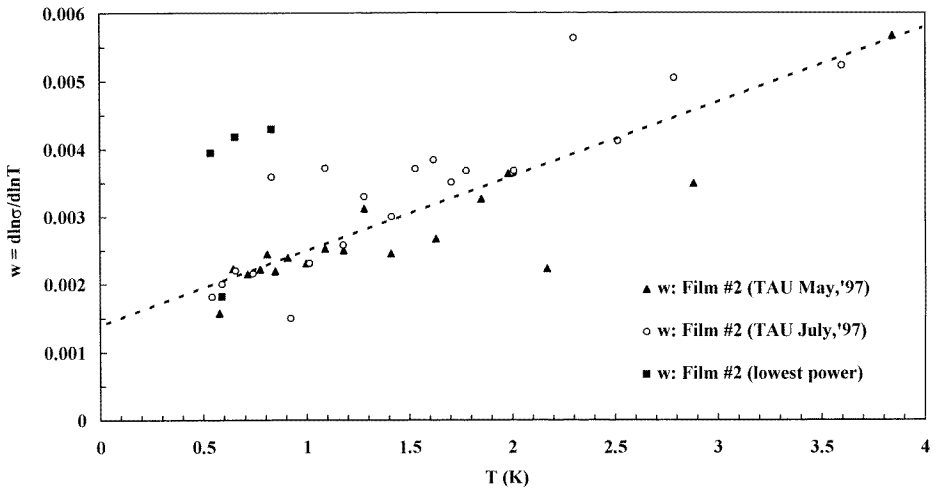

Figure 6. A plot of w, the logarithmic derivative, against temperature for film No 2. Note that w *does not* extrapolate *to zero* as $T \to 0$, thus identifying this film as the *first insulating* film of the series.

temperature T as $w = z + T/T_0$, then *mathematically* the conductivity can be described by $\sigma(T) = CT^z \exp(T/T_0)$. Assuming that T_0 is much greater than the T-values, the exponential can be expanded to yield $\sigma(T) \approx CT^z + DT^{z+1}$; and the conductivity clearly vanishes as $T \to 0$, making the film insulating. A good fit to the conductivity data below 4 K is given by $\sigma(T)/(\Omega^{-1}\ \mathrm{m}^{-1}) = 46\,590(T/\mathrm{K})^{0.0014} \exp(T/910\ \mathrm{K})$. We are not aware of any physical model that would explain this behaviour. However, the w-criterion strongly suggests that film No 2 is *insulating*. Note that film No 2 is more resistive than film No 1.

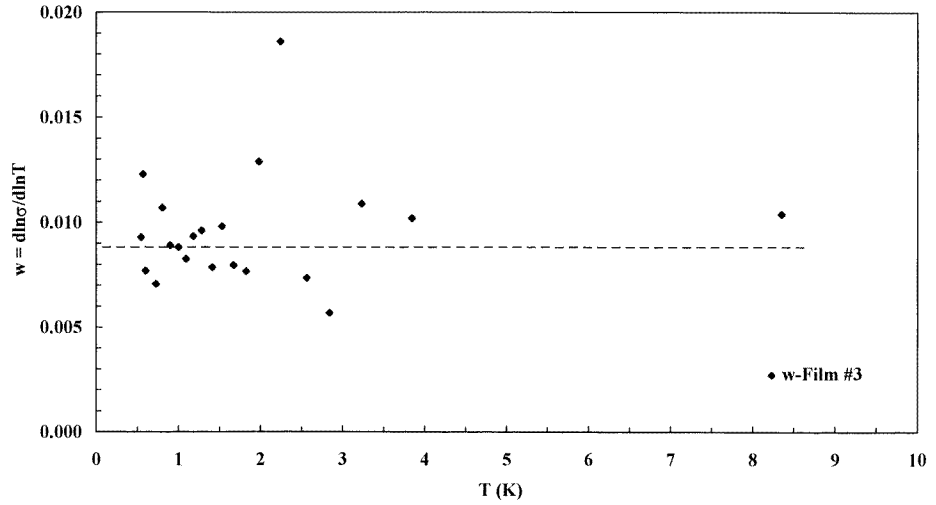

Figure 7. A plot of w, the logarithmic derivative, against temperature for film No 3. w for film No 3 exhibits the 'weakly insulating' *temperature-independent* behaviour implying that $\sigma(T) = CT^z$. This film is clearly insulating.

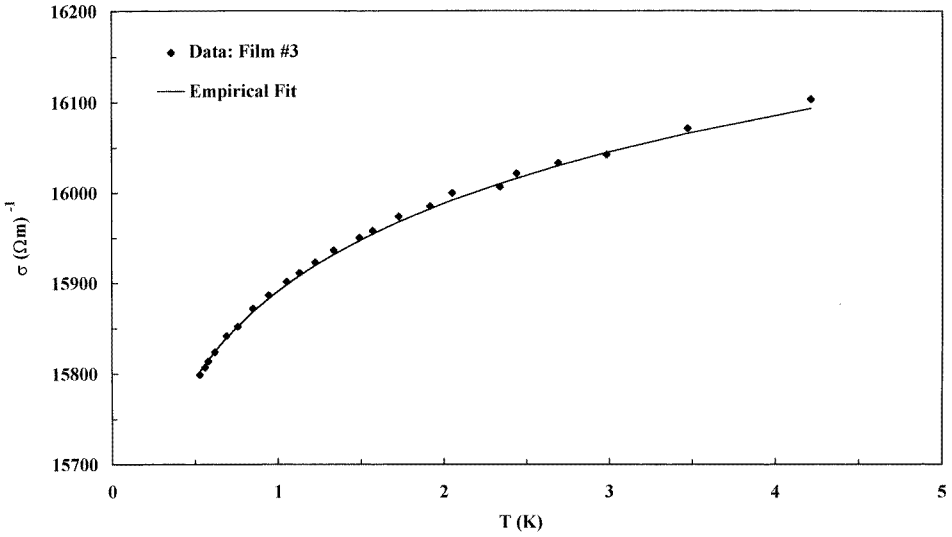

Figure 8. A fit of the empirical insulating conductivity expression $\sigma(T) = CT^z$ to the conductivity data for film No 3. See the text for fitting details.

Film No 3, which is even more insulating and further from the MIT, displays only the 'weakly insulating' behaviour of constant w-values as seen in figure 7; and its conductivity data can be fitted nicely using the insulating empirical expression $\sigma(T)/(\Omega^{-1}\ \mathrm{m}^{-1}) = 15\,890(T/\mathrm{K})^{0.0088}$ as seen in figure 8. Probably, the crossover region for film No 3 to variable-range hopping is in the low-mK region.

In addition, the w-values for film No 42, which is even more insulating, clearly suggest a crossover around 0.1 K from a 'weakly insulating' behaviour at higher temperatures where

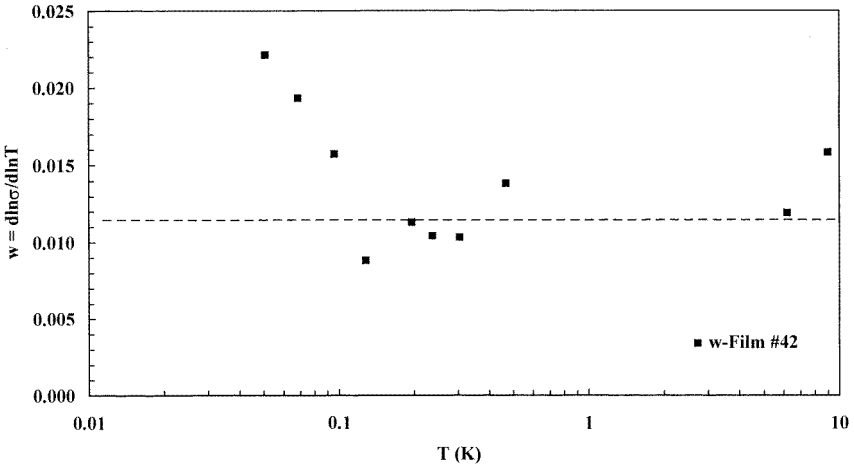

Figure 9. A plot of w, the logarithmic derivative, against temperature for film No 42. w for film No 42 exhibits a crossover at around 0.1 K from the 'weakly insulating' behaviour at higher temperatures where $\sigma(T) = CT^z$ to variable-range hopping behaviour at the lowest temperatures where $\sigma(T) = \sigma_0/\exp(T_0/T)^y$. This film is clearly insulating.

$\sigma(T) \propto (T/\mathrm{K})^{0.11}$ to a variable-range hopping behaviour, indicated by the *increase* of w below 100 mK as seen in figure 9. Unfortunately, we do not have data to sufficiently low temperatures to define the variable-range hopping conductivity law below 0.1 K The behaviour of w in the mK region points to the importance of extending resistance measurements to as low a temperature as possible in order to ascertain whether a film is insulating or metallic.

We found that film No 20, the most insulating film in this series, exhibited the commonly observed crossover in its conductivity from Mott variable-range hopping at high temperatures to Efros–Shklovskii hopping at low temperatures [28].

Thus, the metal–insulating transition lies somewhere between films No 2 and No 1 ($8.7 \times 10^{25}/\mathrm{m}^3 < n < 11 \times 10^{25}/\mathrm{m}^3$). According to figure 3, this would place limits on the parameter $k_F l_e$, namely $4.8 < k_F l_e < 5.5$. Mott's and our suggestions that $k_F l_e \approx \pi$ appear to be a better criterion than $k_F l_e \approx 1$ for locating the metal–insulator transition in this amorphous indium oxide film series. In contrast, Tousson and Ovadyahu suggested that $k_F l_e \approx 0.8$ for amorphous $\mathrm{In}_x\mathrm{O}_y$ in their figure 2 of reference [21]; their small experimental magnitude probably resulted from the lack of resistivity data to sufficiently low temperatures and from a dubious criterion for determining the MIT. In addition, Imry suggests that the critical conductivity at the MIT must be much smaller than $10^6\ \Omega^{-1}\ \mathrm{m}^{-1}$ for amorphous semiconductors [29]; his prediction is consistent with the mean conductivity of 60 000 ($\Omega^{-1}\ \mathrm{m}^{-1}$) for films No 1 and No 2 according to table 1.

The Ioffe–Regel criterion can probably be used successfully on other systems to locate their metal–insulator transitions using room temperature measurements, provided that the Hall voltages can be measured with sufficient accuracy and provided that the Hall constant behaves continuously across the MIT.

Acknowledgments

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