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Hopping transport in an insulating quasicrystal bar of $Al_{70}Pd_{22.5}Re_{7.5}$ near the metal–insulator transition

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Abstract
We have observed that the conductivity $\sigma(T)$ for the $Al_{70}Pd_{22.5}Re_{7.5}$ quasicrystal, with a resistivity ratio $r = R(4.2\text{ K})/R(300\text{ K}) = 13.2$, obeys the variable-range hopping law, $\sigma(T) = \sigma_0/\exp[(T_0/T)]^{\mu}$, in the temperature range between 64 mK and 1.6 K. The hopping exponent μ is extracted to be 0.23, close to the Mott exponent of 1/4, and T_0 is 3.5 K. This insulating behaviour is consistent with the prediction of a previously determined scaling law that bulk $Al_{70}Pd_{22.5}Re_{7.5}$ samples having $r \geqslant 12.8$ will be *insulating*. Large positive magnetoresistances (MRs) were observed in this sample. The percentage change in the MR $= \Delta R(B,T)/R(0,T)$, as high as 185% at $T = 0.11$ K and $B = 17$ T, is the largest value ever reported in Al–Pd–Re QCs, to our knowledge. The difficulties using existing MR theories to explain the MR data for this insulating sample near the metal–insulator transition are discussed.

1. Introduction

The series of Al–Pd–Re quasicrystals (QCs) display the highest resistivity ≈1 Ω cm [1–4] at 4.2 K ever reported amongst various QCs. The high resistivity is due mainly to a wide and deep pseudogap around the Fermi energy E_F [5–7] such that the density of state at E_F, $N(E_F)$, is much smaller than that for other QCs. The electrons located at E_F tend to be localized, possibly owing to quasiperiodicity [8] or to aggregates of clusters [9]. High quality Al–Pd–Re QCs with a wide range of temperature resistivity ratios r's $= \rho(4.2\text{ K})/\rho(300\text{ K}) \approx 1$–280 can be prepared with special annealing procedures [1, 10].

It is known that Al–Pd–Re QCs with $r \ll 13$ are *metallic*; and the variations of their resistance and magnetoresistance (MR) can be explained well by metallic quantum interference theories, namely by the weak localization (WL) theory and by the electron–electron interaction (EEI) theory as demonstrated experimentally [11, 12].

Moreover, Al–Pd–Re QCs with r values greater than 13 exhibit *insulating* behaviour; sometimes their low T conductivity exhibits Mott's variable range hopping (VRH) behaviour [13–17]. In most cases, the temperature interval of the VRH regime is limited to less than a decade, or the conductivity exhibits a tendency to saturate to a constant value at low temperatures. The behaviour of our data is much different. The MR, negative at low magnetic field and positive at high magnetic fields [14, 15], can be described satisfactorily by the forward interference (FI) and the wavefunction shrinkage (WFS) theories developed for a disordered system in the VRH regime [18]. These results clearly show that there is a metal–insulator transition (MIT) in the Al–Pd–Re QCs series fabricated at Tainan. Note that we define the sample as *insulating* if its electrical conductivity *vanishes* at absolute zero.

Knowledge of the physical origins of the MIT in Al–Pd–Re QCs is important for fully understanding the electronic properties of QCs.

In 1996, Lin *et al* [19] found that McMillan's scaling theory of the MIT in disordered systems [20], taking into account localization, correlation and screening, can be applied to *metallic* QCs equally well. That is, the low temperature conductivity $\sigma(T)$ on the *metallic* side of the MIT varies as

$$\sigma(T) = \sigma(0)[1 + (T/\Delta)^{1/2}] = \sigma(0) + mT^{1/2}, \tag{1}$$

where Δ, called the correlation gap, is proportional to the square of the zero-temperature conductivity $\sigma(0)$ and Δ goes to zero at the MIT [19]. This indicates that the EEI plays an essential role in the low T conductivity for quasicrystalline samples near the MIT. Based on equation (1), Wang *et al* [16] performed a series of studies on the low T conductivity of $Al_{70}Pd_{22.5}Re_{7.5}$ QCs with $r \leqslant 18$ and found the extrapolated $\sigma(0)$ obeys the following scaling law:

$$\sigma(0) = \sigma_0(1 - r/r_c)^{\nu}, \tag{2}$$

where the values of σ_0, ν and r_c are $\sigma_0 = 25.6 \pm 3.8\ \Omega^{-1}\ \mathrm{cm}^{-1}$, $\nu = 1.0 \pm 0.15$ and $r_c = 12.8 \pm 0.5$, respectively [15, 16]. r_c is the critical resistivity ratio at which the MIT occurs. Thus, QCs having r's $\leqslant 12.8$ should be *weakly insulating*. Note that the extrapolations are based upon data taken above 1.5 K [16, 19]. The critical exponent $\nu \approx 1$ is also observed in amorphous semiconductors alloyed with transition metals like Nb_xSi_{1-x} [21], Au_xSi_{1-x} [22] and Mo_xGe_{1-x} [23] exhibiting the EEI process.

In order to check the reliability of equation (2) and to gain more insight into the MIT in QCs, we prepared a quasicrystalline sample $Al_{70}Pd_{22.5}Re_{7.5}$ with $r = 13.2$, which according to equation (2), is supposed to be a weak insulator. We now report its low temperature conductivity and MR behaviour.

2. Experimental procedures

Ingots of icosahedral $Al_{70}Pd_{22.5}Re_{7.5}$ QCs were obtained by arc melting a mixture of high-purity Al (99.999 wt%), Pd (99.99 wt%) and Re (99.99 wt%) in a purified argon atmosphere. The ingots were pulled into a bar shape and were sealed in a quartz ampoule, annealed in vacuum at 950 C for 24 h. The samples were then cut into bar shapes with dimensions of $0.9 \times 1.6 \times 5\ \mathrm{mm}^3$. The geometric factor f_g needed to convert resistance to resistivity for this $r = 13.2$ bar sample was $f_g = 0.0403$ cm. Current and voltage leads were attached to the bar with silver paint.

Measurements were made by inserting the bar directly into the mixing chamber of a dilution refrigerator to enhance thermal contact to the cold dilute He^3–He^4 mixture. The dilution

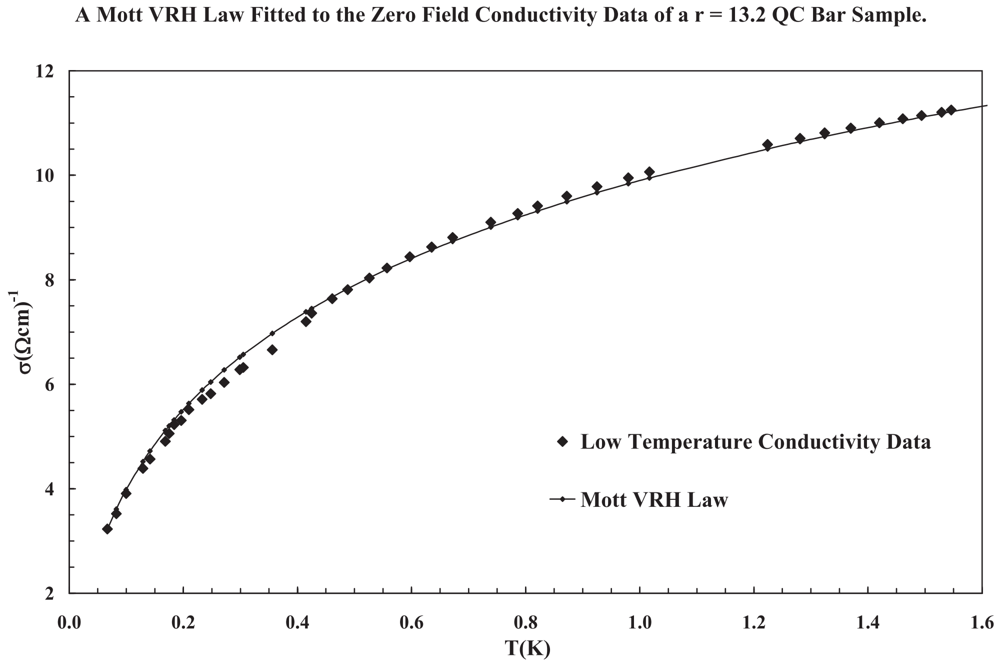

Figure 1. Zero field conductivity data $\sigma(T)$ versus temperature for the $Al_{70}Pd_{22.5}Re_{7.5}$ QC having $r = 13.2$. The full curve represents a Mott VRH fit to the data.

refrigerator at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee, Florida is equipped with a 17.5 T superconducting magnet. Minimum excitation power of 5×10^{-9} W was used to avoid Joule heating of the sample. Hence, a compromise was made between the magnitude of the measured voltage and the background noise. Measurements of the small ac voltages and excitation currents were made using the Stanford Instrument SR830 lock-in amplifiers. We have found that other commercial instruments lead to excessive Joule heating of the sample, as is clearly illustrated in [24].

The NHMFL dilution refrigerator is *not* installed in a shielded room and hence the thermometry and sample responses are very sensitive to external rf heating from the environment. The resistance of the QC was reproducible on a daily basis down to 125 mK. But below 125 mK, data taken only on the weekends were accepted, owing to the very quiet rf background during these days. All measurements below 60 mK were unreliable and were disregarded. Also, long waiting periods of several hours were required to achieve thermal equilibrium between the thermometer and the QC sample, separated from one another. In addition, long time constants of 30–60 s were used on the output of the lock-in amplifier to average the voltage noise fluctuations. Nevertheless, there was considerable scatter in the very low temperature data owing to these experimental noise problems and shielding limitations.

3. Results

3.1. Zero field conductivity—Mott VRH law fit

Figure 1 shows the conductivity $\sigma(T)$ versus temperature between 64 mK and 1.6 K. $\sigma(T)$ is seen to decrease down to the lowest measured temperature *without exhibiting saturation*, at least down to 64 mK.

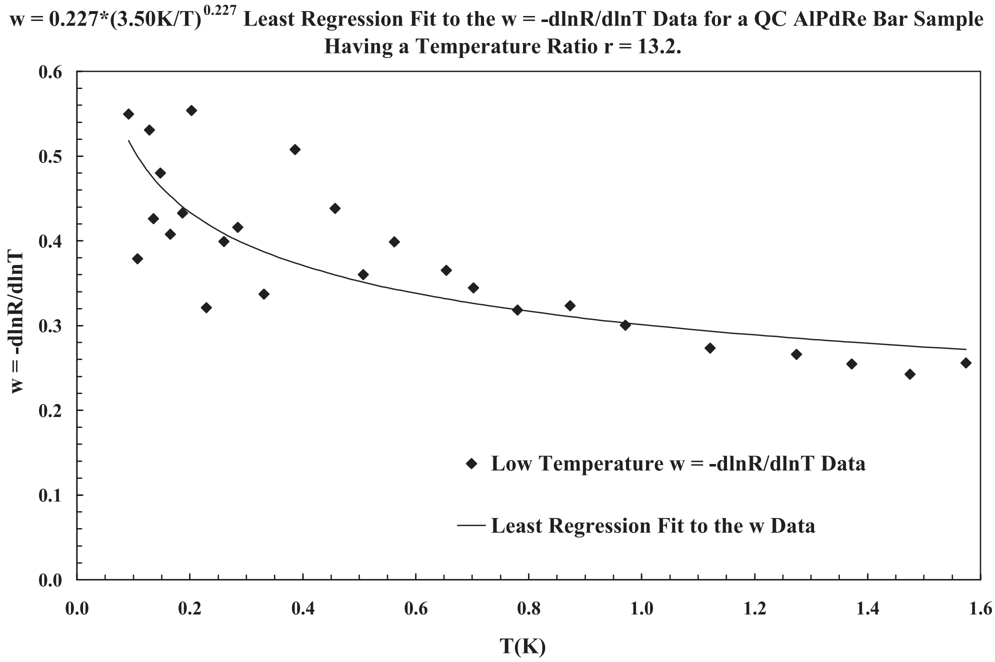

Figure 2. $w = \mathrm{d}\ln\sigma/\mathrm{d}\ln T$ as a function of temperature for the $Al_{70}Pd_{22.5}Re_{7.5}$ QC having $r = 13.2$. The full curve is a least regression fit to the $w = \mathrm{d}\ln\sigma/\mathrm{d}\ln T$ data, which yield values for the Mott characteristic temperature T_{Mott} and the hopping exponent μ.

The function

$$w(T) = \mathrm{d}\ln\sigma/\mathrm{d}\ln T = (T/\sigma)(\mathrm{d}\sigma/\mathrm{d}T) = -(T/R)(\mathrm{d}R/\mathrm{d}T) \tag{3}$$

is extremely useful to characterize whether the sample is insulating or metallic. As detailed in [24], if w either *increases* or is a *constant* with decreasing temperature, then the sample is *insulating*. In contrast, if w extrapolates to *zero*, then the sample is *metallic*. Least regression fits to the w data yielded two of the three fitting parameters needed to fit the theories to the conductivity data. The third fitting parameter in each theory was determined by evaluating a known conductivity value at a given temperature. The following expressions are all in units of $1/(\Omega\ \mathrm{cm})$.

A plot of w against temperature is shown in figure 2. w is clearly seen to *increase* with *decreasing* temperature. This implies that electron transport is via an *activated* hopping process in the sample and $\sigma(T)$ as a function T can be described by a VRH law [25]:

$$\sigma(T) = \sigma_{exp}/\exp[(T_0/T)^{\mu}], \tag{4}$$

yielding

$$w = \mu(T_0/T)^{\mu}. \tag{5}$$

The w data are quite noisy, but this is very characteristic of data taken below 0.5 K, mainly owing to the low dissipation power used in the measurements. A least regression fit to the $w = \mathrm{d}\ln\sigma/\mathrm{d}\ln T$ data, shown by the full curve in figure 2, yielded $\mu = 0.23 \pm 0.015$ and $T_0 \approx 3.5 \pm 0.30$ K. Note that the hopping exponent 0.23 is close to the Mott value of 1/4. In figure 1, the resulting fit (full curve), given by

$$\sigma(T) = 37.3/[\exp(3.5/T)]^{0.23}, \tag{6}$$

is compared to the data and agreement is *good* between the activated fit and the data. *No additional residual* conductivity term was included in the fit. This result is in direct conflict with the findings of Ahlgren *et al* [26].

Mott made an intriguing prediction for the conductivity behaviour for the case when the Fermi energy is centred in the middle of a *deep* pseudogap of the density of states (DOS), as outlined in [25]. Mott speculated that electrons in this deep gap region will be localized and hence the system should be *insulating*, and the conductivity should display a *Mott VRH law* [25]. Smith and Ashcroft have suggested that the strong pseudogap is created by the strong Fermi surface–Brillouin zone interaction [27].

It will be interesting to extend measurements to much lower temperatures, firstly to see if a crossover to Efros–Shklovskii VRH takes place [28, 29], and secondly to see if there is a tendency for the conductivity to eventually saturate, as was observed in Al–Pd–Re QCs with *much higher r* values [26] at liquid helium temperatures.

Interestingly, the experimental value for the exponential prefactor of equation (6), $\sigma_{\exp} = 37.3\ 1\ \Omega^{-1}\ \mathrm{cm}^{-1}$, is of the same magnitude as the prefactor $\sigma_0 = 25.6\ 1\ \Omega^{-1}\ \mathrm{cm}^{-1}$ appearing in the scaling law expression of equation (2). A theoretical explanation is needed for this surprising similarity.

Using $T_{\mathrm{Mott}} \approx 3.5$ K, the localization length a_0 can be estimated [25] from the relation $a_0 = \{18/[k_B N(E_F) T_{\mathrm{Mott}}]\}^{1/3} \approx 310$ Å, where $N(E_F)$ was evaluated from the γ value $\approx$0.1 mJ/(g-atom K^2) determined from specific heat measurements [1]. The ratio of the hopping distance R_{hop} to the localization length a_0, $R_{\mathrm{hop}}/a_0 = 0.375(T_{\mathrm{Mott}}/T)^{1/4}$ is only about 1.1 to 0.5 for 64 mK $\leqslant T \leqslant$ 1.6 K. This indicates that the sample is indeed a weak insulator and is located just below the MIT.

Delahaye *et al* [30] reported that the conductivity for some quasicrystalline ribbons of $Al_{70.5}Pd_{21}Re_{8.5}$ obeys a true Mott VRH law at very low temperatures; that is, $\sigma(0) = 0$ and $\mu = 0.25$ in the temperature range 20–600 mK. They obtained a value of $T_{\mathrm{Mott}} \approx 1$ mK. Their T_{Mott} value $\approx$ 1 mK is much smaller than our value of 3.5 K, indicating that their sample is nearer the MIT. However, their r value $\approx$100 is much larger than our value of 13.2. This is in contrast to our understanding that a sample with a higher r value should be more deeply insulating.

3.2. Zero field conductivity—alternative fits

One might question whether the Mott VRH law yields the best possible fit, compared to other possible models.

First, let us consider the Efros–Shklovskii VRH law where the conductivity follows the activated law with the hopping exponent of 1/2 [31]:

$$\sigma(T) = \sigma_{\exp}/\exp[(T_{ES}/T)^{1/2}]. \qquad (7)$$

A least regression fit yielded the following expression:

$$\sigma(T) = 15.7/\exp[(0.22/T)^{1/2}]. \qquad (8)$$

The fit to the data is illustrated in figure 3 by the broken line and the ES fit is clearly poorer than the Mott fit. According to Castner [28], the crossover temperature T_{cr} to the ES regime should be approximately 80 times smaller than the Mott temperature or about 44 mK, a temperature that is lower than our lowest measurement temperature. Thus, we should not observe ES VRH in our ‘high temperature’ conductivity data.

There are theoretical suggestions that a simple temperature power law fit should describe the conductivity for an *insulating* sample as pointed out by Poon [32], namely

$$\sigma(T) = cT^z. \qquad (9)$$

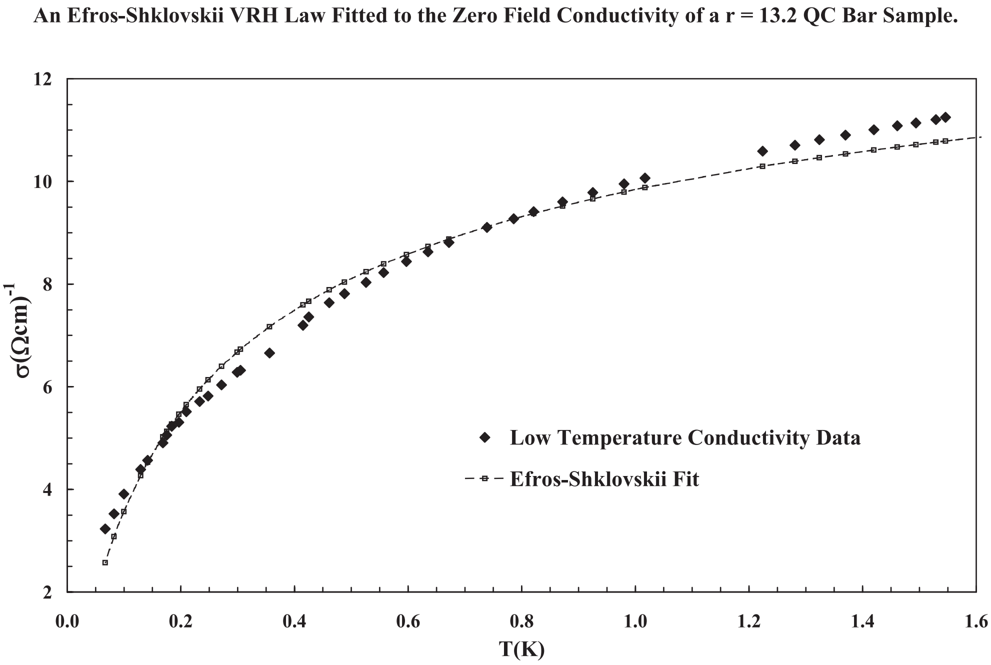

Figure 3. Conductivity data compared to an Efros–Shklovskii VRH law fit.

According to Poon, this simple power law arises if the wavefunctions decay according to the power law $\psi(r) \sim r^{-\beta}$ [13, 32]. For this special case, values of w should be a *constant* as a function of temperature, equal in value to z, the power law exponent. Inspecting the w versus T curve in figure 2, the w values are *very roughly constant* at a value of approximately 0.38. A least regression fit of equation (9) to the data yielded

$$\sigma(T) = 9.92T^{0.39} \tag{10}$$

and the fit is shown in figure 4 by the dashed-dotted curve. This simple power law fit is inferior to the Mott VRH fit. Note that our exponent of 0.39 differs from Poon's suggested value of 0.5 [13, 32].

However, the reader might question whether this QC bulk sample is truly insulating and whether the conductivity might better be described by a *metallic* WL law. Let us momentarily consider that the QC is now *metallic* and that we can fit *metallic* theories to the conductivity data. Note that this assumption is most likely incorrect, since there is no possibility that the w data in figure 2 can be extracted to zero as $T \rightarrow 0$ K, characterizing *metallic* behaviour.

According to the EEI theories of Altshuler and Aronov [33] and Lee and Ramakrishnan [34], the conductivity in *bulk 3D metallic* samples should display a $T^{1/2}$ law near the MIT:

$$\sigma(T) = \sigma_{0,EEI} + CT^{1/2}. \tag{11}$$

A least regression fit to the data gives

$$\sigma(T) = 1.96 + 7.83T^{1/2}. \tag{12}$$

The EEI fit to the data is illustrated by the broken–crossed line in figure 5; the fit is poor.

Some bulk *metallic* samples very close to the MIT exhibit a $T^{1/3}$ law, as demonstrated by Newson and Pepper [35] and discussed by Imry [36], using the diffusion equation.

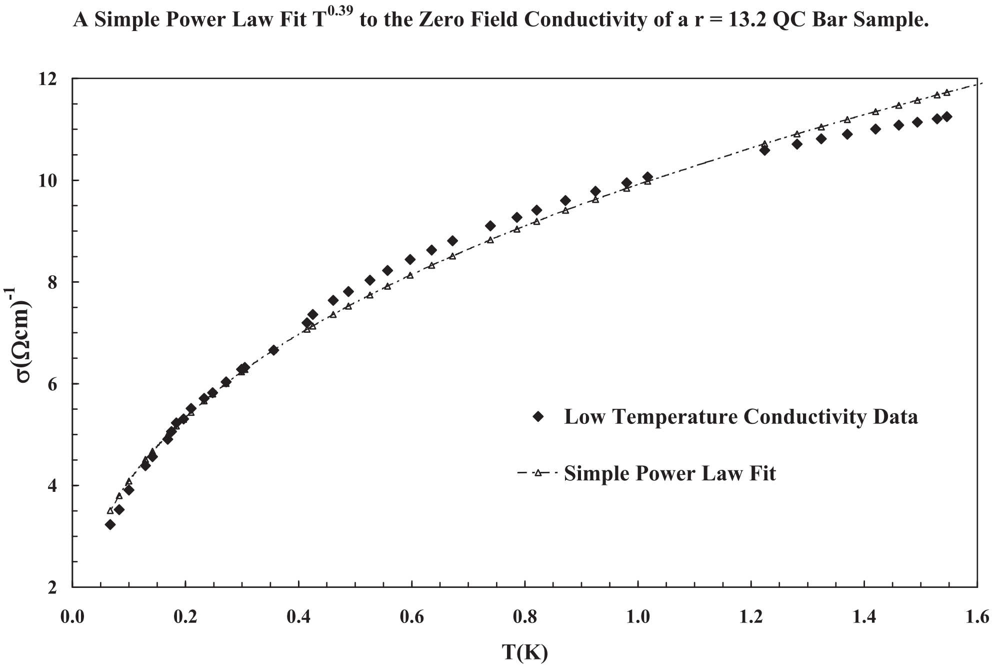

Figure 4. Conductivity data compared to a simple power law fit $T^{0.39}$.

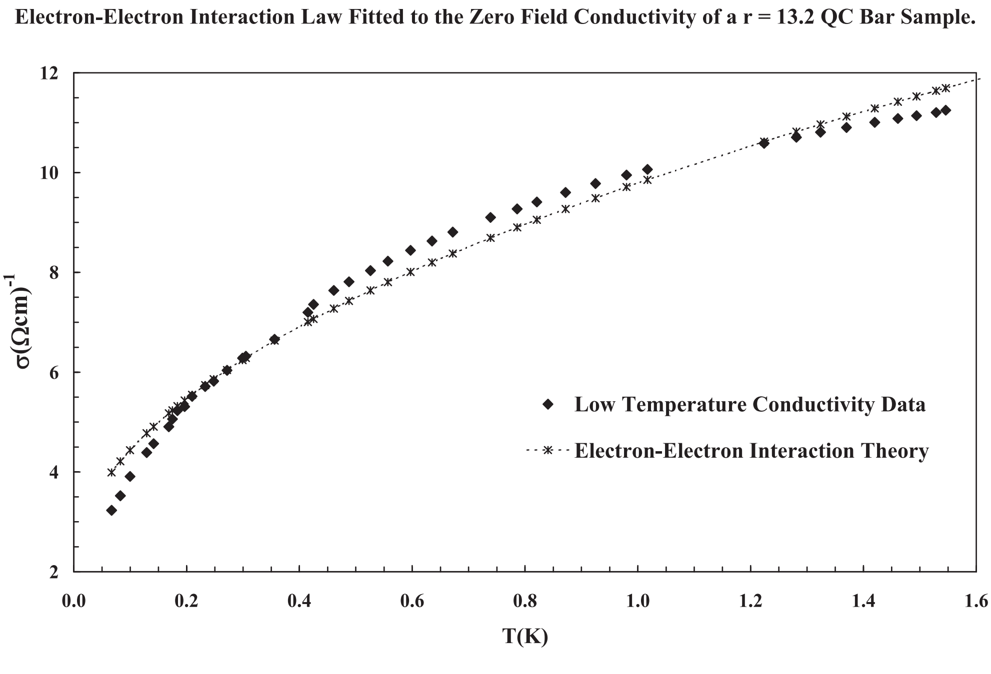

Figure 5. Conductivity data compared to an EEI fit, assuming that the QC is *metallic*.

Thus, the conductivity crosses over from a $T^{1/2}$ law at higher temperatures to a $T^{1/3}$ law at very low temperatures:

$$\sigma(T) = \sigma_{0,\mathrm{dif}} + CT^{1/3}. \tag{13}$$

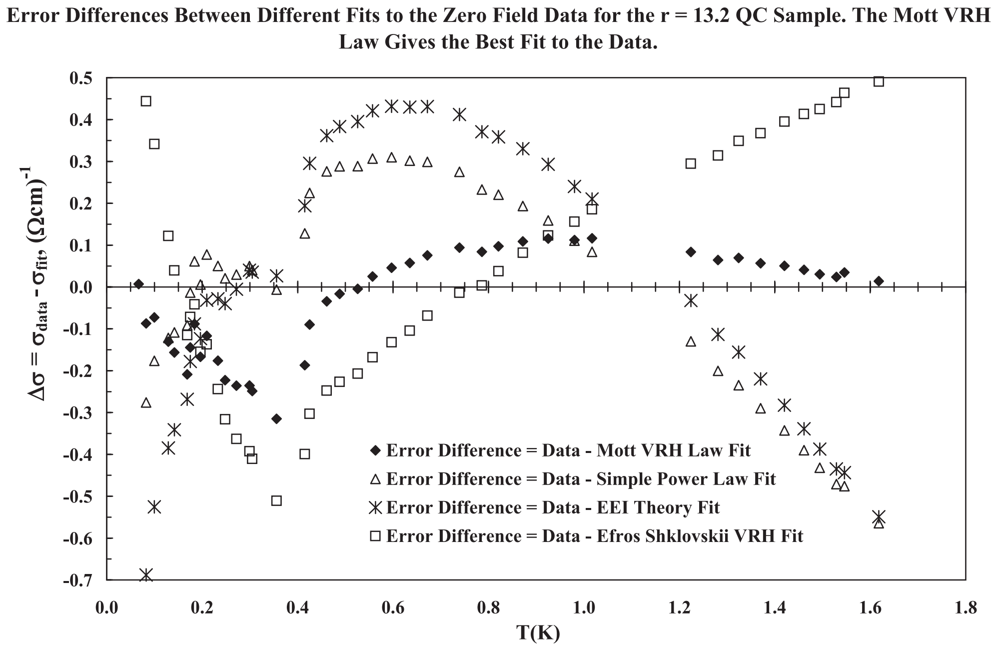

Figure 6. Comparison between various fits and the data, illustrating the 'goodness' of each fit. The Mott VRH fit clearly yields the superior fit.

The least regression fit yielded the following results:

$$\sigma(T) = -0.826 + 9.92T^{1/3}. \tag{14}$$

The fit to the data is excellent, *mathematically*, but *completely unphysical* owing to the *minus sign* for the zero-temperature conductivity term $\sigma_{0,\mathrm{dif}}$. For this reason, we have not plotted this diffusion model fit.

The most convincing test that the conductivity follows a Mott VRH law is a plot of the differences between the actual data values and the values predicted by the different models. These plots are shown in figure 6. It is clear that the Mott VRH model yields the superior fit.

Our results clearly show that the scaling law of equation (2) correctly predicts that the $r = 13.2$ QC bar sample is weakly insulating. Interestingly, for quasicrystalline Al–Pd–Re *films* [24], the value of r_c appears to be somewhat smaller than 8.7.

3.3. Zero field conductivity at high temperatures

The conductivity data above 10 K exhibits a linear power law dependence on temperature as shown in figure 7. The full line is a fit given by

$$\sigma(T) = 17.4 + 0.59T. \tag{15}$$

Janot has predicted the linear T dependence [37], motivated by measurements made by Pierce *et al* [1].

4. Magnetoresistance data

Figure 8 shows the normalized MR ratio $r_{\mathrm{MR}} = R(B, T)/R(0, T)$ versus magnetic field B. Unlike the $Al_{70}Pd_{22.5}Re_{7.5}$ QCs with $r \geqslant 18$, where the MR appears negative at low fields

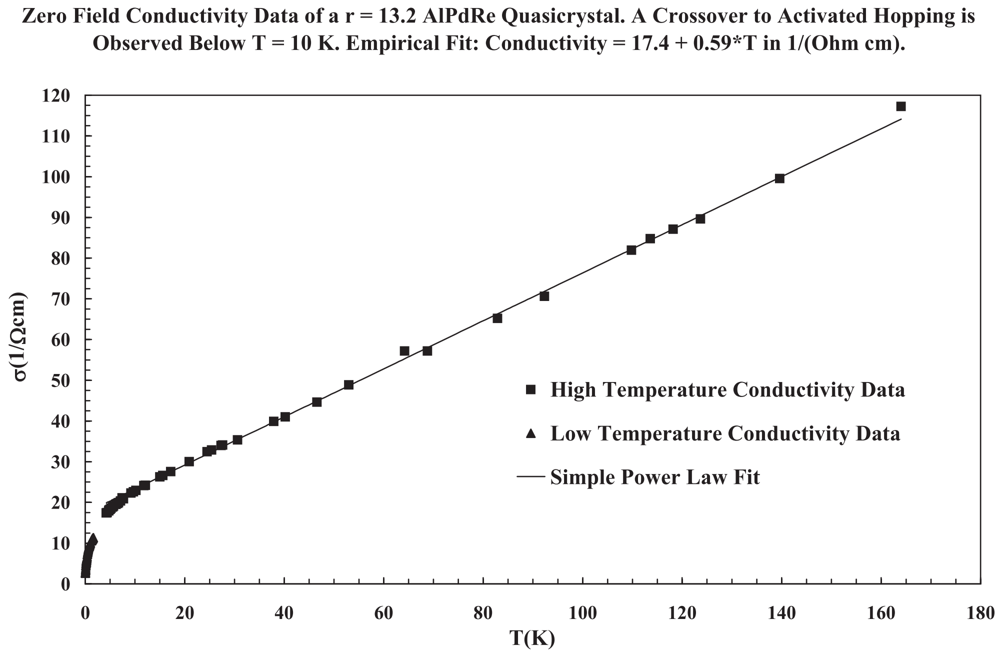

Figure 7. Zero field conductivity data taken at high temperatures. The full line represents a simple temperature power law fit, $\sigma \propto T^{1.0}$.

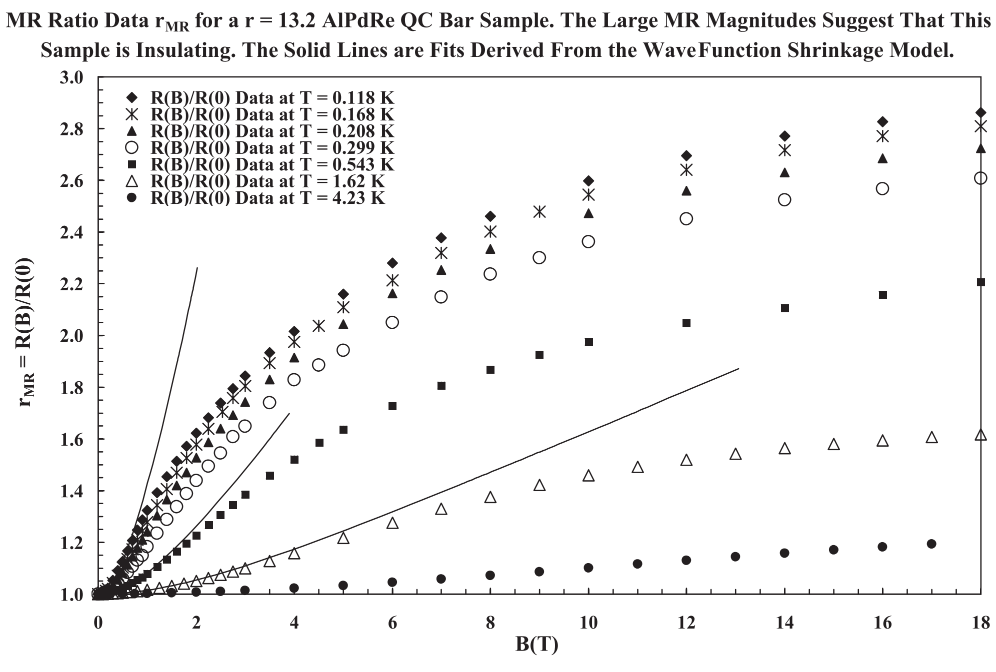

Figure 8. MR ratio $r_{MR} = R(B)/R(0)$ data at different temperatures. The full curves represent fits of the WFS theory.

and becomes positive at high fields [15, 18], the observed MR is only positive within the measured field range. At $T = 0.11$ K and $B = 17$ T, the value of r_{MR} is about 2.85 or $\Delta R(B, T)/R(0, T) \approx 185\%$; to our knowledge, this is the largest value ever reported in $Al_{70}Pd_{22.5}Re_{7.5}$ QCs.

There is one major problem when analysing the MR data for this *weakly insulating* sample. We are not aware of any theory to explain the MR for *weakly insulating* 3D samples. There are several theories valid for *strongly* insulating samples. We define *strongly insulating* samples by the criterion $r_{hop}/a_0 = 0.385(T_{Mott}/T)^{1/4} > 1$ when the sample is in the Mott VRH regime or by the criterion $r_{hop}/a_0 = 0.25(T_{ES}/T)^{1/2} > 1$ when the sample is in the Efros–Shklovskii VRH regime. Here r_{hop} is the average hopping distance between occupied and vacant sites and a_0 is the localization length. For our QC sample having $T_{Mott} \approx 3.5$ K, values for $r_{hop}/a_0 = 0.385(T_{Mott}/T)^{1/4}$ range between 1.05 and 0.45 for the temperature range between 0.064 and 1.64 K. Again these small values for r_{hop}/a_0 support the claim that this sample is *weakly insulating*.

One is strongly tempted to fit these MR data using the WL theory and the EEI theory valid for *metallic* samples located just above the MIT. But this is a dangerous procedure since these theories predict that the zero field conductivity should follow a temperature power law dependence, which is not our case, nor can these magneto*conductance* theories predict *such large values* for the MR ratios which we observe at the lowest temperatures without introducing non-physical fitting parameters. We also note that the 3D WL at large fields predicts a $B^{1/2}$ dependence upon the magnetoconductivity, $\Delta\sigma_{WL}(B, T)$, which we do not observe experimentally [38, 39].

One could also consider the MR fitting procedure used by Srinivas *et al* [40] on their *strongly insulating* QCs, where they found that their QCs exhibit the Efros–Shklovskii VRH in the conductivity. This fitting procedure was originally introduced by Schoepe [41]. But, as we have demonstrated above, the conductivity of our QC sample does *not* follow the ES VRH law and thus we have not adopted their procedure.

Recently, we have shown in [18] that, for Al–Pd–Re QCs with large r values that are *well inside* the VRH regime, the negative MR is due to the FI effects [42], while the positive MR is mainly caused by the WFS effect [41, 43, 44]. Both of these theories assume that the sample is *strongly localized*, either in the Mott or ES regimes. We will now apply these two theories to the *weakly localized* regime, having no better alternative models available.

The FI theory describes that the interference among many paths between two sites is predominantly destructive; thus, suppression of the destructive interference by a magnetic field results in a negative MR [42]. It was found that, as the sample's r value is decreased, the negative MR reduces rapidly and cannot be detected experimentally for $r < 18$ [15]. Consequently, the FI effects should be small and can be ignored in this sample.

The WFS theory considers the contraction of the electronic wavefunction at the impurity centres in a magnetic field, which leads to a reduction in the hopping probability between two sites and therefore to a positive MR [43, 44]. Based on the WFS theory, Schoepe [41] derived an expression for the MR ratio $R(B, T)/R(0, T)$ valid for the entire field regime. The expression for $R(B, T)/R(0, T)$ can be written as

$$R(B, T)/R(0, T) = \exp\{\xi_c(0)[\xi_c(B)/\xi_c(0) - 1]\}. \qquad (16)$$

$\xi_c(B)/\xi_c(0)$ is the normalized hopping probability parameter and $\xi(0) = (T_{Mott}/T)^{1/4}$ for the Mott VRH case. Tabulated values of $\xi_c(B)/\xi_c(0)$ as a function of B/B_c for the Mott VRH case, as well as for the ES VRH case, are given in [45]. The only fitting parameter in equation (16) is a characteristic field B_c, given for the Mott VRH case by

$$B_c = [6\hbar/(ea_0^2)][T_{Mott}/T]^{1/4}, \qquad (17)$$

where a_0 is the localization length. Theoretical fits are shown in figure 8 by full lines and are poor, which is not surprising. The value of a_0 calculated from the MR fit is $a_0 \approx 600$ Å, which is twice as large as the 310 Å value estimated from the density of states $N(E_F)$ and the Mott temperature T_{Mott}.

A positive MR contribution based upon a spin mechanism was proposed by Kurobe and Kamimura [46]. But, as pointed out by Vaknin *et al* [47], this formalism is valid again only in the strongly insulating regime. An interesting model based upon hopping MR induced by Zeeman splitting has been suggested by Matveev *et al* [48] and expanded by Meir [49]. In this model, blocking of hopping from a singly occupied site to another singly occupied impurity site takes place owing to spin polarization. Saturation of the resistivity with field is predicted. More theoretical work is needed to explain the MR data in the *weakly insulating* region.

In summary, this $Al_{70}Pd_{22.5}Re_{7.5}$ QC with $r = 13.2$ exhibits Mott VRH behaviour at low temperatures, consistent with the scaling law prediction that this sample is insulating. We do not have a complete understanding of the localization mechanism that leads to the Mott VRH behaviour of the conductivity at low temperatures. Nor do we have a theoretical understanding of the physics in the crossover region, which separates the simple linear temperature power law behaviour of the conductivity at high temperatures from the activated Mott behaviour of the conductivity at low temperatures.

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