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Abstract

Resistivity and magnetoresistance measurements have been performed on insulating icosahedral AlPdRe quasicrystal (QC) bar samples. At temperatures in the range 300 K $\geqslant T \geqslant$ 50 K, the resistivities follow a simple inverse temperature law: $\rho(T) = \rho_0/T^{(1.0\pm0.1)}$. Below 1 K, the resistivity of a *weakly insulating* sample exhibited a simple inverse temperature law where $\rho(T) = \rho_0/T^{0.33}$ and not an activated variable-range hopping (VRH) law. *Strongly insulating* samples exhibit saturation of their resistivities to finite values as $T \to 0$ K. These saturation resistivity values are believed to arise from the presence of a second metallic phase located within the quasicrystal's structure. By extrapolating the measured resistivities at 22 mK to absolute zero, the saturation conductivity values were estimated at $T = 0$ K and subtracted from the conductivity data points. These 'corrected' data, corresponding only to the QC phase, were found to follow activated VRH laws, having hopping exponents y that vary in the range $0.18 \leqslant y \leqslant 0.43$. The activated VRH behaviours are observed *only* below 1 K. The magnetoresistances (MRs) of these samples are also anomalous. The MRs can be explained by including contributions from both the saturation conductivity values and from the QC MR ratios, estimated using the wavefunction shrinkage model.

1. Introduction

Since the first fabrication of the icosahedral i-AlPdRe quasicrystals (QC) structure by Tsai *et al* [1] in 1990, there has been continuous interest and study of the transport properties of this intriguing and challenging material. In 1993 both the groups of Pierce *et al* [2] and of Akiyama *et al* [3] demonstrated the high resistivities in this QC material. In 1994, Pierce *et al* [4] showed that certain heat treatment procedures enhanced the *insulating* behaviour of the resistivity. The Stockholm group of Rapp in a series of publications [5–9] illustrated the *saturation* behaviour to finite constant resistivity (or conductivity) values as $T \rightarrow 0$ K for the AlPdRe QC structure. Guo and Poon [10] introduced the important concept that the saturation behaviour arises from the presence of a second *metallic* phase whose conductivity σ_{sat} 'shorts out' the *insulating* conductivity of the QC primary phase σ_{QC}; they proposed the expression:

$$\sigma_{\text{meas}}(T) = \sigma_{\text{sat}}(T) + \sigma_{\text{QC}}(T), \tag{1}$$

where $\sigma_{\text{meas}}(T)$ is the measured conductivity of the sample. Guo and Poon assumed that the QC conductivity σ_{QC} follows an activated variable-range hopping (VRH) law and that it *vanishes* as $T \rightarrow 0$ K. Rodmar *et al* [7, 11] have presented experimental evidence, based upon SEM scans, for the presence of the second phase. Our data are consistent with the two-phase picture of the QC samples, but contradict published claims of VRH laws dominating above 1 K. Details are now presented.

2. Experimental procedures

Ingots of icosahedral (i) $Al_{70}Pd_{22.5}Re_{7.5}$ QCs were fabricated by arc melting a mixture of high purity Al, Pd and Re in a purified argon atmosphere; these samples were fabricated in Professor S T Lin's laboratory in Tainan, Taiwan. The ingots were sealed in a quartz ampoule and annealed in vacuum at $950\,^\circ$C for 24 h [12, 13]. Some ingots were heat treated for a second time at $600\,^\circ$C for 24 h to enhance their insulating behaviours [4]. The samples were then cut into bar shapes with dimensions of approximately $0.9 \times 1.6 \times 5$ mm^3. Current and voltage leads were attached to the bars with silver paint.

The QCs were initially investigated in Professor J-J Lin's laboratory in Hsinchu, Taiwan. The initial results were exciting and interesting, but additional measurements were needed at lower temperatures. The QCs were transferred to a top loading Janis He^3 refrigerator, model HE-3-TLSL, recently installed at the National High Magnetic Field Laboratory (NHMFL). This refrigerator reaches 0.23 K and is equipped with a 17.5 T superconducting magnet. Upon analysing the data, it became clear that much lower temperatures were needed to extract out the zero temperature saturation conductivity σ_{sat} for each QC sample. The QC samples were placed into an Oxford Instrument top loading dilution refrigerator, model TLM-400, with an 18 T superconducting magnet at the NHMFL. Precautions were used to minimize the Joule heating and rf pickup in the QC samples. Sufficiently long intervals between measurements were used to assure that the samples, the temperature sensors and the cooling liquid were in thermal equilibrium.

X-ray diffraction patterns have been performed on very similar insulating AlPdRe QC samples showing that the alloys can be identified having face-centred icosahedral lattices [12]. Surprisingly, there was no change of the diffraction pattern for a sample that was subjected to the second annealing step at $600\,^\circ$C [12]. Interestingly, there are also additional weak diffraction peaks present that might be associated with the second phase; these peaks are nicely illustrated in figure 1 of [12].

Analysis of the resistivity data is based upon the w technique [14–16]:

$$w = \mathrm{d} \ln \sigma / \mathrm{d} \ln T = (T/\sigma)(\mathrm{d}\sigma/\mathrm{d}T). \tag{2}$$

This technique will identify the sample as either *insulating* or *metallic* and will also reveal the temperature dependence of the conductivity.

Samples may be classified as being either metallic or insulating. *Metallic* 3D samples always display finite resistivities or non-zero conductivities at absolute zero. In contrast, purely *insulating* single phase 3D samples exhibit infinite resistivities or *zero* conductivities at absolute zero.

The conductivity of a 3D *metallic* single phase sample at sufficiently low temperatures can be described by the power law expression:

$$\sigma(T) = \sigma(0) + CT^{z}; \tag{3}$$

$\sigma(0)$ is the conductivity at $T = 0$, arising perhaps from static defects and impurities, C is a prefactor and z is the exponent of the temperature power law. Equation (3) might approximate the conductivity contribution from the 3D electron–electron interaction (EEI) theory [17] and/or from the 3D weak localization (WL) theory [18] or the combination of both processes. Note that the terms in equations (1) and (3) have completely different physical interpretations for the two different cases of insulating and metallic samples.

Strongly insulating single phase samples exhibit activated hopping conductivities, which can be described by the activated VRH expression in zero magnetic field:

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

here T_0 is the characteristic temperature, y is the hopping exponent and σ_0 is the prefactor and a fitting parameter.

If the insulating sample exhibits an activated conductivity law according to equation (4), then

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

and $w(T)$ will *diverge to infinity* as the temperature approaches absolute zero. A least regression fit of the $\log(w)$ versus $\log(T)$ data will determine both the hopping exponent y and the characteristic temperature T_0. Five of the more *strongly insulating* QCs displayed this activated VRH law below 0.3 K.

Moreover, there is a special *weak insulating* case for which $\sigma(0)$ is set to zero in equation (3). In this case, the conductivity follows a simple power law:

$$\sigma(T) = CT^{z}. \tag{6}$$

Equation (2) yields

$$w = z, \tag{7}$$

where z is the power law exponent, independent of temperature.

For 3D *metallic* samples exhibiting slowly decreasing conductivities with decreasing low temperatures, then according to equation (3), $w(T)$ should *extrapolate to zero* as $T \rightarrow 0$ K. From our previous experience, the typical magnitudes of the ws for a *metallic* sample are *small* and of the order of 0.01 at temperatures below 0.1 K [19].

Thus, both the temperature behaviour and the magnitudes of w clearly identify the sample as either metallic or insulating. Any finite value of w at $T = 0$ K implies insulating behaviour for a single phase material. We now apply the w technique to the QC data.
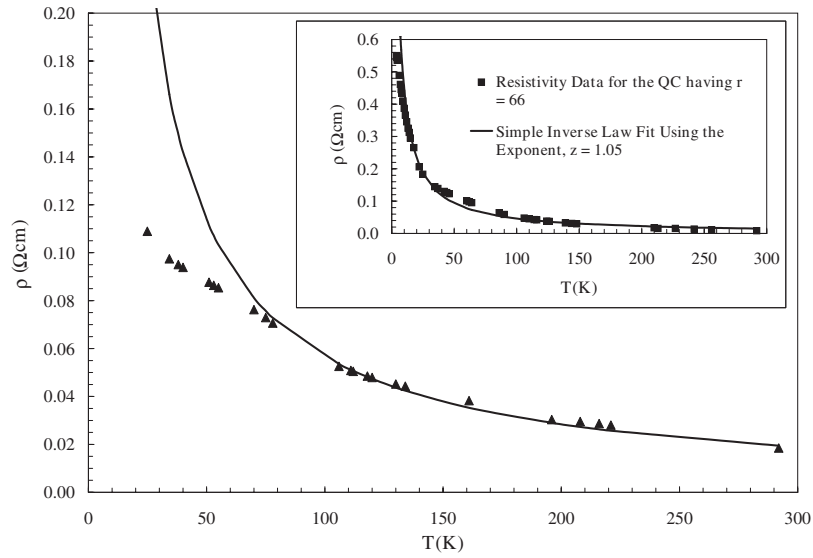
Figure 1. High temperature resistivity data (triangles) for an AlPdRe QC having $r_T = 9.3$. The fit (solid curve) is a simple inverse temperature law where $\rho(T) = 5.68/T^{1.00}$ in Ω cm. The inset shows data (squares) for the QC having $r_T = 66$; the fit (solid curve) is $\rho(T) = 5.88/T^{1.05}$ in Ω cm.

3. Experimental results

3.1. Weakly insulating QC sample

The resistivity data for the weakly insulating $r_T = 9.3$ bar sample are shown in figures 1 and 2; here $r_T = R(4.2\,\text{K})/R(292\,\text{K})$ is the temperature resistance ratio. This sample has a geometric factor f_g of 0.0384 cm, used to convert resistances to resistivities. The *high* temperature data in figure 1 follow closely a simple inverse temperature law given by $\rho(T) = 5.7/T^{1.0}$ Ω cm down to 70 K. This dependence has been suggested by Janot [20]. There is a crossover to another insulating behaviour below 50 K.

The *very low* temperature resistivity data for the $r_T = 9.3$ QC are surprising, as seen in figure 2. There is little indication of saturation tendencies except below 80 mK. In fact, it is not possible to estimate a 'saturation' conductivity value at $T = 0$ K from this data; much lower temperature measurements are required. The signature of saturation clearly appears in the w data below 80 mK, shown in the inset of figure 2, where the ws start to drop to zero owing to σ_{sat}. The unexpected behaviour, however, is the constant temperature independent behaviour of the ws above 80 mK. The ws fluctuate about the average value of about 0.33, indicated by the *solid* line in the inset of figure 2. According to equation (7), this implies a simple inverse temperature law for the resistivity, which can be fitted nicely according to the expression $\rho(T) = 0.26/T^{0.34}$ in Ω cm (or $\sigma(T) = 3.85T^{0.34}$ Ω^{-1} cm^{-1}) as shown in figure 2 over the temperature range. Note that *no* additional saturation conductivity σ_{sat} term is used in the fit. We are not aware of any group that has observed this temperature dependence in their insulating QCs.

In an important publication Zvyagin suggested that the conductivity very close to *both sides* of the metal–insulator transition follows a $T^{1/3}$ dependence [21]; his model uses the 3D scaling theory of the conductivity and non-optimal hopping, characterized by smaller hopping
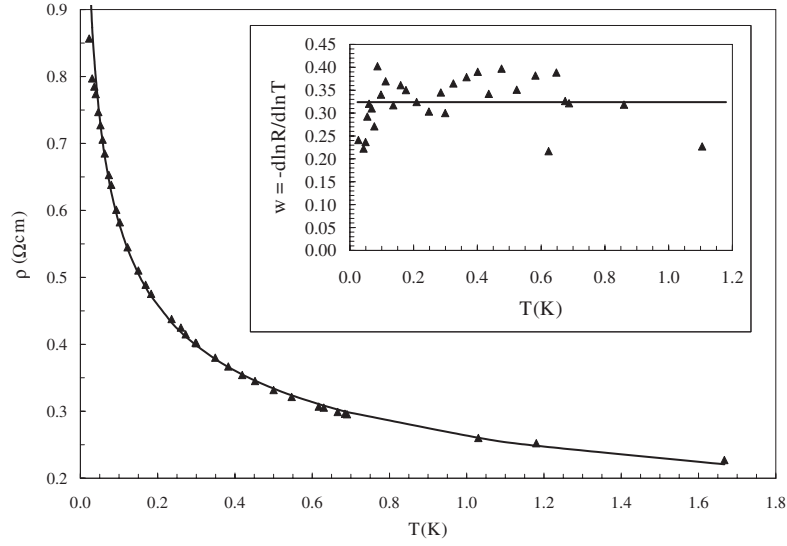
Figure 2. Low temperature resistivity data for an AlPdRe QC having $r_T = 9.3$. The solid curve is a simple inverse temperature law fit: $\rho(T) = 0.263/T^{0.34}$ in Ω cm. No saturation conductivity term σ_{sat} was included. The inset shows the temperature independent behaviour of $w \approx 0.33$, implying a simple inverse law for $\rho(T)$.

distances and energy compared to the conventional variable-range hopping problem [21]. Newson and Pepper extended his calculations [22] and predicted that the prefactor C of the $T^{1/3}$ term is:

$$C = (e^2/\hbar)(G_c)^{2/3}[N(E_F)k_B]^{1/3}, \tag{8}$$

with $G_c = 2/(3\pi^3) = 0.0215$ being the dimensionless critical conductance [23]. $N(E_F)$ is the density of states (DOS) at the Fermi energy of the *insulating* QC. The expression $[N(E_F)k_B T]^{-1/3}$ is the optimal hopping length L to nearest thermally accessible sites, according to Zvyagin [21].

In order to evaluate equation (8), we need an estimation for the DOS in the pseudogap, $N(E_F)$, for the *insulating* AlPdRe QC. For the *metallic* AlPdRe QC, we estimate the metallic DOS $N_{metal}(E_F) \approx 3.6 \times 10^{46}$ states/(J m^3). We used the specific heat prefactor $\gamma = 0.28$ mJ mol^{-1} K^{-2} [24, 25], $\rho_{density} \approx 4.6$ g cm^{-3}, 1 mol ≈ 57 g, Avogadro's number $= 6.02 \times 10^{23}$ atoms mol^{-1} and the conversion relation $N(E_F) = 0.422\gamma$ [26] where the units for $N(E_F)$ are in states/(eV atom).

However, we must estimate the DOS of the *insulating* AlPdRe QC and not the *metallic* DOS. Owing to the pseudogap at E_F of the DOS in the *insulating* material, Mott suggested that an *insulating* sample must have a DOS which is a factor of at least *four times smaller* than the metallic DOS value [27]. Esther Belin-Ferré has summarized DOS measurements about E_F made on bulk QCs and on surfaces of the QCs [28]. According to Janssen and Fasolino [29], the surface DOS should be identical to the bulk DOS. Tunnelling measurements on the surface of i-AlPdRe by Davydov *et al* [30] and by Escudero *et al* [31] show deep dips in the DOS at E_F, which are about a factor of *eight* times smaller than the DOS values that are distantly spaced from E_F. Thus, we estimate $N_{insul}(E_F) \approx 4.5 \times 10^{45}$ states/(J m^3). Inserting these values for $N_{insul}(E_F)$ and G_c into equation (8) yields $C \approx 7.5$ Ω^{-1} cm^{-1} K$^{-1/3}$, in surprisingly good agreement with the experimental value of 3.85 Ω^{-1} cm^{-1} K$^{-1/3}$. Thus, the predictions of Zvyagin and of Newson–Pepper describe well the weakly insulating conductivity [21, 22].
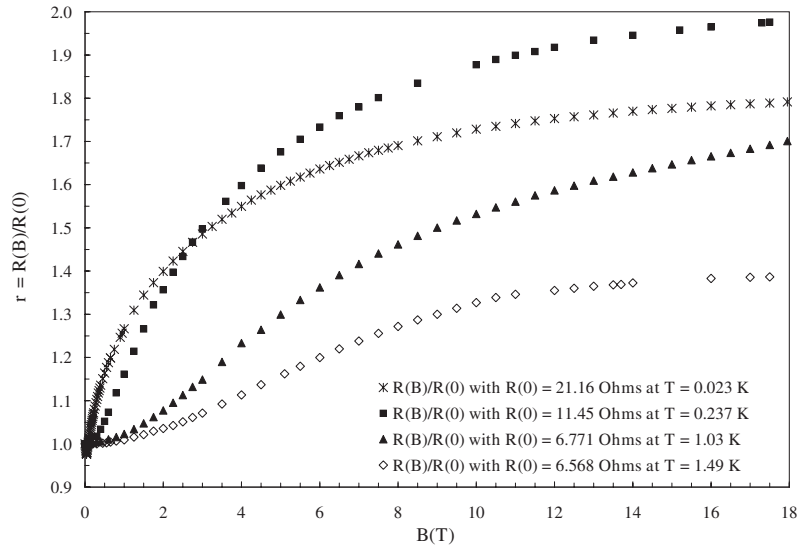
Figure 3. MR ratios r at different temperatures for the AlPdRe QC having $r_T = 9.3$. The large magnitudes suggest insulating behaviour. The cross over behaviour of the $T = 0.023$ K data to smaller values above $B = 3$ T suggests a weak saturation behaviour. A MR theory is needed for this *weak insulating* regime.

The MR ratio data, $r = R(B)/R(0)$, at different low temperatures are shown in figure 3 for the $r_T = 9.3$ QC. Note that the *positive* ratios approach the value of two at high fields and low temperatures, strongly suggesting insulating behaviour. Such large values cannot be explained by the WL theory [18, 32] or by the EEI theory [17, 33], valid only for *metallic* samples [34]. We are *not* aware of *any* MR theory for *weakly insulating* samples [34]. There is one anomalous behaviour, appearing in the 23 mK MR data of figure 3. This is the 'cross over' to smaller ratio values above 3 T as compared to the 230 mK data; we propose an explanation below.

3.2. Strongly insulating QC sample

We now summarize the transport behaviour of a more insulating AlPdRe QC having a temperature ratio $r_T = 66$ and a geometric factor $f_g = 0.0629$ cm. The high temperature resistivity data are shown in the inset of figure 1, where again a simple inverse resistivity law, $\rho(T) = 5.9/T^{1.05}$ in Ω cm, gives a good fit to the data from room temperature down to 20 K. This QC shows strong signs of saturation to a constant saturation value of about $\rho_{sat}(T = 0, B = 0) \approx 3.075$ Ω cm, as can be deduced from the low temperature resistance data of figure 4. It would be desirable to extend the data down to 10 mK to better ascertain the saturation conductivity value $\sigma_{sat}(T = 0, B = 0) = 1/\rho_{sat}(T = 0, B = 0) \approx 0.325$ Ω^{-1} cm^{-1}. Note that these resistivity data are still increasing significantly even down to 22 mK, in strong contrast to behaviours reported by other groups [5–9]. Thus the magnitudes for σ_{sat} appear to vary widely between different QC fabrication groups; the σ_{sat} values from Professor S-T Lin's group seem to be smaller than those of other groups.

The w plot reveals important features of the resistivity behaviour, as shown in the inset of figure 4. First, if one calculates the ws directly from the raw data, then the ws extrapolate nicely to zero, suggesting the presence of the saturation conductivity term σ_{sat}; we have not shown this
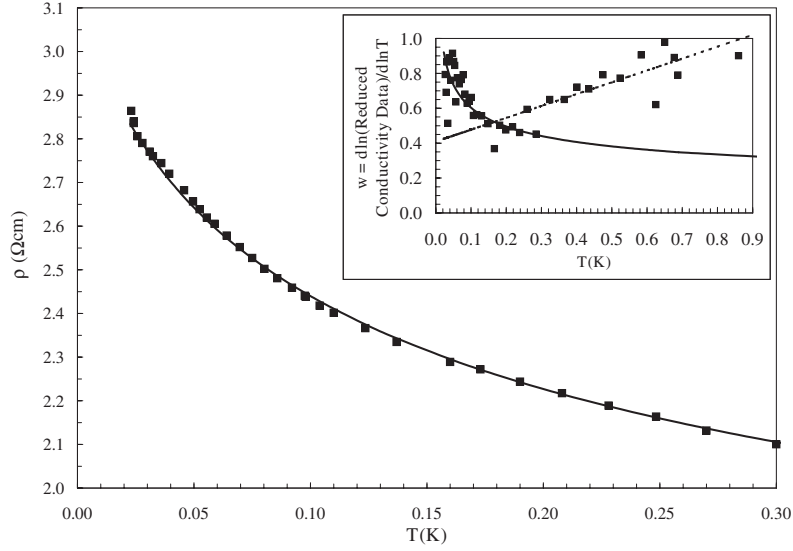
Figure 4. Low temperature resistivity data for an AlPdRe QC having $r_T = 66$. The solid curve includes both the saturation conductivity term and the VRH law conductivity, using a hopping exponent $y = 0.285$. By extrapolating $T \to 0$ K, the saturation conductivity value is estimated to be $0.325 \ \Omega^{-1} \ cm^{-1}$. The inset shows w derived using the 'reduced' data (conductivity data–saturation conductivity value). There are two regions: at low Ts below 0.25 K there is activated hopping (solid curve); at intermediate Ts there is a 'linear T' behaviour of w (dashed line).

plot, since it yields no additional information. However, when one subtracts off the saturation conductivity value $\sigma_{sat}(T = 0, B = 0) \approx 0.325 \ \Omega^{-1} \ cm^{-1}$ from all the measured conductivity data points $\sigma_{meas}(T, B = 0)$ to find the QC conductivity contribution, $\sigma_{QC}(T, B = 0)$, namely

$$\sigma_{QC}(T, B = 0) \approx \sigma_{meas}(T, B = 0) - \sigma_{sat}(T = 0, B = 0), \tag{9}$$

and then calculates the ws using these estimated σ_{QC} values, one obtains the surprising results shown in the inset of figure 4. We observed that the ws decrease linearly with temperature from 0.9 down to 0.3 K, exhibit a minimum around 0.25 K and then *increase* below 0.25 K down to 0.022 K. The presence of this increase is exciting, since it signifies *activated* hopping. A least regression fit of the $\log(w)$ versus $\log(T)$ data below 0.25 K yielded values for the hopping exponent of $y = 0.285$ and the characteristic temperature $T_0 = 1.39$ K. Thus, the best empirical fit to the measured resistivities, including both the contributions of the constant saturation term and the activated VRH term, becomes:

$$\rho(T) = 1/[0.325 + 0.705 \exp{-(1.39 \ K/T)^{0.285}}], \tag{10}$$

in units of Ω cm. The fit is excellent, as seen in figure 4, and extends over one decade of temperature. Thus it appears that the resistivity of the QC follows a general activated law but *not* a Mott VRH law with $y = 1/4$ [35]. If we could eliminate the presence of the second phase by improved sample preparation, then we would anticipate that the QC resistance will extrapolate to infinity as $T \to 0$ K. Refer to [16] for a discussion on the 'linear w versus T' dependence observed above 0.3 K.

Four other insulating QCs exhibited activated hopping, having VRH fitting parameters that are summarized in table 1. There are *no* agreements between our hopping exponents and those reported in [6]. However, there is one report of a hopping exponent $y = 0.23$, close to the Mott 1/4 value [36].
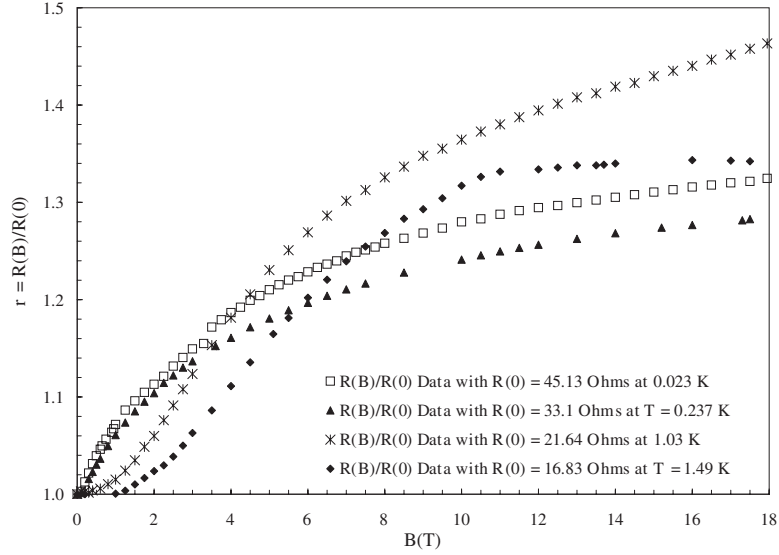
Figure 5. MR ratios r at different temperatures for an AlPdRe QC having $r_T = 66$. The MR behaviour is anomalous at the lowest temperatures and at the highest fields.

Table 1. VRH fitting parameters extracted from the w fits.

$r_T = R(4.2\ \text{K})/R(292\ \text{K})$ ratios of the QCs	Hopping exponent y	Characteristic temperature T_0 (K)
21.7	0.187	1300
19.3	0.216	118
66	0.285	1.39
116	0.356	1.16
>136	0.436	0.22

The MR behaviour for this $r_T = 66$ QC is shown in figure 5. The behaviour is anomalous at high fields since the MR values are *smaller* at the two lowest temperatures as compared to the larger values at the two highest temperatures. The same behaviour also appeared in figure 3 for the $r_T = 9.3$ QC. These behaviours are not predicted theoretically.

We suggest that the anomalous MR behaviour also arises from the presence of the second phase.

We can express the MR ratios $r = R(B)/R(0)$ at a fixed measuring temperature T_{fix} theoretically as:

$$r(B, T_{\text{fix}}) = \sigma_{\text{meas}}(B = 0, T_{\text{fix}})/[\sigma_{\text{sat}}(B, T_{\text{fix}}) + \sigma_{\text{QC}}(B, T_{\text{fix}})]. \tag{11}$$

The denominator term is identical to equation (1) where we assume that the total conductivity contribution is composed of the sums from the metallic second phase term and from the insulating primary QC phase term, but now these two terms are field *dependent*. The numerator term, $\sigma_{\text{meas}}(B = 0, T_{\text{fix}})$, is simply a normalization factor. It is related to the measured zero field resistance $R(0)$ value at the fixed measured temperature T_{fix} through the geometric factor f_g of the sample—namely, $\sigma_{\text{meas}}(B = 0, T_{\text{fix}}) = 1/[f_g R(0, T_{\text{fix}})]$. Note that in *zero* field, the denominator term must be *equal* to the numerator term to yield a ratio value of 1. At $B = 0$ T,

we can now estimate the magnitude for σ_{QC}, since we have measured σ_{meas} and have estimated σ_{sat} via the resistivity extrapolation to $T = 0$ K.

We inquire whether we can predict the *field dependences* for the two denominator terms. Consider first the insulating QC conductivity σ_{QC} behaviour, in particular below 0.3 K where the majority of our QCs display activated VRH laws. Recently, the MR ratio behaviour $r = R(B)/R(0)$ has been predicted using the 'wavefunction shrinkage' (WFS) model [37], provided that the hopping exponent y and characteristic temperature T_0 appearing in the VRH expression are known. Recall that both these fitting parameters have been extracted from the w data. There is only one additional fitting parameter, B_c, that must be estimated directly from fits to the MR ratio data [37]. At low temperatures and high fields, the model predicts ratios r that increase by one to two orders of magnitude. This large increase of $R(B)$ results from the electronic wavefunctions being distorted from 'spherical' shapes in zero field to 'cigar-type' patterns in large fields; the distortion results in small values for the overlap integrals and small probabilities of the electron hopping to an unoccupied site. Thus the *QC* conductivity term σ_{QC} becomes negligible at high magnetic fields.

By contrast, the metallic conductivity term σ_{sat} of the second phase has *little* field dependence and only a *small* temperature dependence. We propose that the metallic second phase is composed of *narrow metallic* ribbons or wires threading through the QC structure. We assume that we can apply the 2D quantum transport corrections to these ribbons, namely the 2D WL theory [38, 39] and the 2D EEI theory [40, 41]. Assuming that the 2D theories can be applied to the ribbons, we know from many experimental results on thin metallic films [19] that the MR of the metal can be described nicely using only the WL theory and that the magnetic field effect is *small*—typically a *1 to 2% increase or decrease* of the zero field resistance in strong fields, depending on whether the film has *strong* spin–orbit (from the Pd or Re) or *weak* spin–orbit interaction (from the Al). Thus, to first approximation, σ_{sat} can be treated as field independent. Also the temperature dependence of σ_{sat} is weak and of the order of a few per cent change [19]. The zero field metallic conductivity increases slightly at finite temperatures owing to the electron–electron interaction correction. Thus, to first order, σ_{sat} can also be treated as temperature independent and approximately equal to its extrapolated value at $T = 0$ K.

If we now consider the behaviour of the two conductivity terms in strong magnetic field, the QC conductivity contribution will tend to zero, leaving only the second phase term to provide the conducting path. Thus, we predict that the MR ratios *should saturate* at high fields and low temperatures to small ratio values given approximately by:

$$r = \sigma_{meas}(B = 0, T_{fix})/[\sigma_{sat}(B = 0, T = 0)]. \qquad (12)$$

The saturation tendency of the MR data is seen in figure 5. In figure 6, we show a fit of this model including both the wavefunction shrinkage (WFS) term and the saturation conductivity term (solid curve) and a second fit excluding the saturation term (dashed curve) to the MR ratio data taken at $T = 0.23$ K. Only one fitting parameter that appears in the WFS model, namely $B_c = 0.9$ T, was used; the value for B_c was chosen to optimize the fit to the low field MR ratio data. We have *not* included the 2D weak localization correction to σ_{sat} since we do not know the chemical composition of the ribbons and wires and hence the strength of the spin–orbit scattering.

We have not made MR measurements at higher temperatures above 1 K. Other groups have published high quality data in this higher temperature region [42, 43].

In conclusion, if we accept the presence of a second metallic conducting phase, then this phase is responsible for the saturation behaviours observed in both the resistivity and MR measurements on insulating AlPdRe QCs. Below 0.3 K, the QC phase generally exhibits an
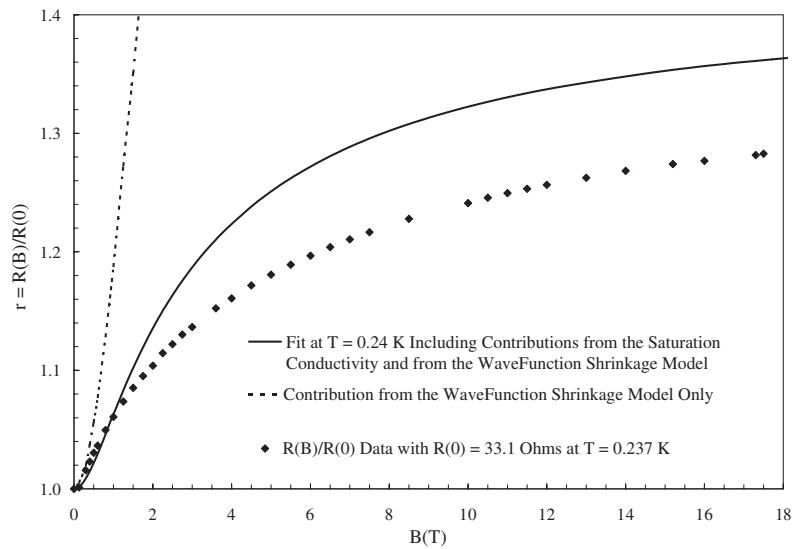
Figure 6. Magnetoresistance ratios at $T = 0.24$ K for an AlPdRe QC having $r_T = 66$. The fit (solid curve) includes contributions from the wavefunction shrinkage model ($y = 0.285$, $T_0 = 1.39$ K and $B_c = 0.9$ T) and the saturation conductivity term (0.325 Ω^{-1} cm^{-1}). The dashed curve is the wavefunction shrinkage fit, *excluding* the contribution of the saturation conductivity term.

activated VRH hopping law in its conductivity. Both the resistivity and MR ratio data can be fitted well taking into account the contributions from both phases.

Acknowledgments

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