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Electron transport and magnetic properties of the icosahedral Al–Pd–Re quasicrystals

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Abstract. The temperature dependence of the conductivity of Al–Pd–Re quasicrystals can be described by a power law, that is, $\Delta\sigma(T) \propto T^{\alpha}$; α is different in low- and high-temperature regimes for high-quality samples. At high temperatures, the conductivity is found to be closely related to the inverse Hall coefficient. The magnetic susceptibility χ_M of Al–Pd–Re quasicrystals is negative. At low temperatures the magnitude of χ_M is seen to decrease quite rapidly with increasing temperature. $\chi_M(T)$ between 2 and 400 K can be described well by the following relation: $\chi_M(T) = \chi_0 + aT^{-\alpha} + bT^2$. The possible origins of each of the terms are discussed in this paper.

1. Introduction

Much attention has been paid to the discovery of the thermodynamically stable quasicrystals (QC) Al–Cu–(Fe, Ru, Os) [1] and Al–Pd–Mn [2] with a face-centred-icosahedral ordered structure, which possess exotic electronic properties [3–6]. Their electron transport properties are found to be very sensitive to the structural quality of the icosahedral phase as well as the fine adjustment of the alloy composition and thermal treatment conditions. It is, in general, believed that the QCs with better quality have higher resistivities, and the high resistivities of these stable QCs are mainly due to (1) the existence of a pseudogap at the Fermi level and (2) the localization tendency of electrons near the Fermi level [6]. The former effect reduces the electron density of states at the Fermi level and is also considered to be the chief reason for the stability of the quasicrystalline structure; the latter reduces the diffusion constants of the electrons. A striking feature is that, despite the fact that the QCs have very high resistivity values [5–7], their conductivities at low temperature can be well described by the effects of weak-localization (WL) and electron–electron interactions (EEI) originally developed for disordered systems [7–11].

Recent studies of i-Al–Pd–Re alloys revealed that this series of i-QCs have the highest resistivity ever reported for QCs [12–15]. The highest values of $\rho(4.2 \text{ K}) \cong 1.5 \Omega$ cm and 2.0 Ω cm are reported for i-Al_{70.5}Pd₂₁Re_{8.5} [13] and for i-Al₇₀Pd₂₀Re₁₀ [15], respectively. These values of $\rho(4.2 \text{ K})$ are about one order of magnitude larger than those for the i-Al–Cu–Fe [16], i-Al–Cu–Ru [17] and i-Al–Pd–Mn [3] systems, and are of the same order of

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magnitude as the ones for doped semiconductors well inside the insulating region of the metal-insulator transition (MIT) [18]. However, the results for the temperature dependence of the conductivity σ obtained by different groups are inconsistent. For example, Honda *et al* [15] found that the conductivity behaviour of i-Al₇₀Pd₂₀Re₁₀ cannot be interpreted in terms of either the theory of WL or conventional variable-range hopping (VRH), but on the basis of measurements of optical conductivity, Basov *et al* [19] showed that i-Al₇₀Pd₂₀Re₁₀ is a semiconductor and its electrical conductivity at low temperature can be attributed chiefly to the VRH mechanism. These contradictory results clearly indicate that there is still no theory which can explain satisfactorily the electric conduction process at low temperatures in i-Al–Pd–Re QCs.

It is known that the magnetic susceptibility of high-quality i-Al–Cu–Fe and i-Al–Pd– Mn with Mn content lower than 8 at.% exhibits diamagnetic behaviour [16, 3]. Since high-quality i-Al–Pd–Re alloys are easily made, these i-alloys provide us with a good opportunity to study their intrinsically magnetic properties. Therefore, in this work, we will not only reinvestigate the electronic transport properties of i-Al₇₀Pd_{22.5}Re_{7.5} but also study the magnetic properties of this alloy.

2. Experiments

Ingots of icosahedral Al₇₀Pd_{22.5}Re_{7.5} and Al_{70.5}Pd₂₂Re_{7.5} alloys were obtained by the arc melting of a mixture of high-purity Al (99.999 wt%), Pd (99.99 wt%), and Re (99.99 wt%) in a purified argon atmosphere. To ensure complete mixing, each ingot was turned upside down and remelted several times in the arc-melting process. The ingots were pulled into a long bar shape and then were sealed in a quartz ampoule, annealed in vacuum at a temperature between 900 °C and 980 °C for 24 hours, and quenched into the ice-water. X-ray diffraction spectra were measured using a rotating-anode x-ray generator (Cu K α , 50 kV, 200 mA) with a graphite (002) monochromator. The resistivity was measured in the temperature range between 4.2 and 300 K using a Linear Research LR-400 AC resistance bridge (~15.9 Hz) with a PC/AT-based automatic data-acquisition program. A measuring current I_{RMS} of 0.1 mA was used. The bar-shaped samples for this measurement have dimensions $\sim 0.9 \times 1.6 \times 9$ mm³. Both the cooling and the warming rates in the cycle measurement were controlled at about 2 K min⁻¹, which is slow enough to ensure that the resistivity $(\rho(T))$ curves measured during both the cooling and warming processes can overlap each other. The Hall coefficient was measured from 30 K to 300 K with a DC current of 8 mA under a magnetic field of 5 T. The temperature stability during each measurement was $\pm 0.1-0.5$. A SOUID magnetometer was used to measure the susceptibility in the temperature range from 2 K to 400 K and in an applied field of 1 T, and the M-H curves at 5 K, 77 K, and 300 K under a magnetic field in the range 0-5.5 T.

3. Results and discussion

The bottom and top panels of figure 1 show the x-ray diffraction patterns for samples $Al_{70.5}Pd_{22}Re_{7.5}$ (APR4) and $Al_{70}Pd_{22.5}Re_{7.5}$ (APR1a) annealed at 950 °C and 940 °C, respectively, for 24 h and then subsequently quenched into the ice-water. The middle panel of figure 1 displays the x-ray diffractogram for the sample $Al_{70}Pd_{22.5}Re_{7.5}$ (APR1b) prepared like sample APR1a at first and then subjected to a further annealing at 600 °C for 2 h. Samples APR1a and APR1b, while not the same sample, are cut from the same ingot. All of the peaks in figure 1 can be indexed using the Elser scheme [20], but only a few



Figure 1. X-ray diffraction patterns for the samples APR1a ($Al_{70}Pd_{22.5}Re_{7.5}$), APR1b ($Al_{70}Pd_{22.5}Re_{7.5}$), and APR4 ($Al_{70.5}Pd_{22}Re_{7.5}$). The conditions of the heat treatment are described in the text.

Table 1. The conditions of the heat treatment and the values of the physical parameters $\rho_{4.2 \text{ K}}$, $\rho_{4.2 \text{ K}}/\rho_{300 \text{ K}}$, and $k_F l$ for i-Al–Pd–Re QCs.

Composition	Annealing temperature (time)	$\rho_{4.2 \text{ K}}$ ($\Omega \text{ cm}$)	<i>р</i> 4.2 к∕ <i>р</i> 300 к	$\begin{array}{c} k_F \times 10^{-2} \\ (m^*/m) \end{array}$
APR1a (Al ₇₀ Pd _{22.5} Re _{7.5})	940 °C (24 h)	0.15	13.8	5.2
APR1b (Al ₇₀ Pd _{22.5} Re _{7.5})	940 °C (24 h)	0.44	46.8	1.8
+	- 600 °C (2 h)			
APR2 (Al ₇₀ Pd _{22.5} Re _{7.5})	980 °C (24 h)	0.10	11.4	8.9
APR3 (Al ₇₀ Pd _{22.5} Re _{7.5})	940 °C (24 h)	0.09	7.2	7.9
APR4 (Al _{70.5} Pd ₂₂ Re _{7.5})	950 °C (24 h)	0.06	7.4	13.8

indexed peaks are shown. It is clearly seen from figure 1 that the alloys can be identified as having face-centred-icosahedral (FCI) lattices. Figure 2 and figure 3 reveal the resistivity ρ and the conductivity σ as functions of temperature, respectively.

The value of the resistivity ρ at 4.2 K and the resistivity ratio $\rho(4.2 \text{ K})/\rho(300 \text{ K}) = r$ for all of the measured samples with almost the same nominal composition are listed in table 1. $\rho(4.2 \text{ K})$ is seen to vary from 0.06 Ω cm to 0.44 Ω cm. The observed variation of $\rho(4.2 \text{ K})$ for these samples is primarily due to the difference in both the actual composition



Figure 2. The resistivity ρ as a function of temperature for the samples APR1a, APR1b, APR2, APR3, and APR4.

and annealing conditions. But appropriate annealing conditions can alter the values of $\rho(4.2 \text{ K})$ and r very significantly. For example, $\rho(4.2 \text{ K})$ and the resistivity ratio r for the sample APR1a (annealed at 940 °C for 24 h) are 0.15 Ω cm and 14, but become 0.44 Ω cm and 46 for sample APR1b (annealed at 940 °C for 24 h with an additional annealing at 600 °C for 2 h). This confirms the finding by Pierce *et al* [13] that the preannealing of i-Al-Pd-Re alloys at high temperatures around 940 °C followed by a further annealing at low temperatures around 600 °C can lead to a large increase in $\rho(4.2 \text{ K})$ and r. It must be emphasized here that the drastic change in $\rho(4.2 \text{ K})$ and r in i-Al-Pd-Re QCs prepared with an additional low-temperature annealing is quite uncommon because it was not observed for i-Al-Pd-Mn QCs by us.

As seen in figure 3, all of the measured conductivities increase with increasing temperature. This is a common feature for nonmagnetic i-alloys like i-Al–Cu–(Fe, Cu) alloys [21, 22]. We have attempted to interpret the conductivity data below 30 K for Al₇₀Pd_{22.5}Re_{7.5} (APR1a and APR1b) by means of the theories of EEI and WL including spin–orbit interaction, and found that although the theoretical calculation can fit the experimental results fairly well, the inelastic scattering time τ_i and spin–orbit relaxation time τ_{so} obtained are two to three orders of magnitude larger than those obtained for the i-alloys [23]. Magnetoresistance measurements on the i-Al_{70.5}Pd₂₁Re_{8.5} [24] and i-Al₇₀Pd₂₀Re₁₀ alloys [15] reveal that the variation of the resistance with magnetic field is quite significant and



Figure 3. The conductivity σ as a function of temperature for the samples APR1a, APR1b, APR2, APR3, and APR4.



Figure 4. Power-law plots of the conductivity for sample APR1b. The inset shows the powerlaw variation of this sample in the temperature range 4.2–50 K. Both solid lines are fits with $\sigma(T) = \sigma_0 + cT^{\alpha}$.

complicated, but these results cannot be simply explained by the WL theory, either.

The condition for the validity of the WL and EEI theories is that $k_F l > 1$, where k_F is the Fermi wave number and *l* the mean free path. We estimate the values of $k_F l$, following reference [5], and list the values in table 1. It is clearly seen from the table that these two theories break down for most of the samples studied unless the ratio of the tangential effective mass to the free-electron mass $m_t^*/m \approx 10^2$ —but this seems quite impossible. The above analyses clearly show that the localization effects in i-Al–Pd–Re QCs are beyond the weak-localization regime. An attempt to employ the conventional VRH theory to explain the low-temperature conductivity of Al–Pd–Re QCs also fails. This may imply that the samples are not insulators yet.

For nonmagnetic i-alloys, the relation between σ and T may be expressed as $\sigma(T) =$

 $\sigma_0 + \Delta\sigma(T)$, where σ_0 depends on the composition and the structural quality of the samples, and $\Delta\sigma(T)$ is found to be nearly independent of the composition [25]. We can see from figure 3 that for APR2, APR3, and APR4, $\Delta\sigma(T)$ is approximately proportional to *T* throughout the whole measured temperature range, but the experimental σ -*T* data for APR1a and APR1b with lower conductivity do not follow $\Delta\sigma(T) \propto T$; instead, it is found that $\Delta\sigma(T) \propto T^{\alpha}$ with α determined to be 0.974 ± 0.003 (4.2–50 K) and 1.289 ± 0.002 (100– 300 K) for APR1a; and 1.191 ± 0.003 (4.2–50 K) and 1.398 ± 0.004 (100–300 K) for APR1b. The result for sample APR1b is shown in figure 4.



Figure 5. The Hall coefficient R_H as a function of temperature for the samples APR1a and APR1b.

Figure 5 plots the Hall coefficients R_H of APR1a and APR1b against temperature from 30 K to 300 K. R_H is positive for both APR1a and APR1b over the measured temperature range. For T < 50 K, the value of R_H for APR1a is larger than that for APR1b, both values drop roughly to the same value between 75–200 K, and then the former tends to decrease more slowly above 200 K. Unlike that for APR1b, the R_H observed by Pierce *et al* [13] for the Al_{70.5}Pd₂₁Re_{8.5} sample (also prepared with an additional low-*T* annealing) is positive at low temperature, but it becomes negative above 40 K.

The discrepancy in both their and our R_H -data may be due to the difference between the Fermi energies of the two samples with different compositions. Since, as demonstrated by the theoretical calculations of Fujiwara *et al* [26] for the crystalline approximant α -AlMn (a simulation of α -AlMnSi), the magnitude and the sign of R_H are very sensitive to the location of the Fermi level inside the pseudogap, where the density of states consists of very fine spiked peaks with a peak width of only about 0.01–0.02 eV. Thus, for a sample with a Fermi level at certain particular locations, its R_H can quite possibly change sign as the temperature varies.

The effective carrier concentration $n_{\text{eff}} = 1/eR_H$ and the Hall mobility $\mu_H = \sigma R_H$ as functions of temperature are shown in figure 6. The above relation between n_{eff} and



Figure 6. The carrier concentration n_H and Hall mobility μ_H as functions of temperature for samples APR1a and APR1b.

Table 2. The values of the parameters χ_0 , a, α , and b deduced from the fit of the χ_M -data to the equation $\chi_M(T) = \chi_0 + aT^{-\alpha} + bT^2$.

Composition	χ_0 (10 ⁷ emu g ⁻¹)	$a (10^7 \text{ emu g}^{-1} \text{ K}^{\alpha})$	α	b (10 ¹³ emu g ⁻¹ K ⁻²)
APR1a	-5.84 ± 0.11	7.57 ± 0.08	0.38 ± 0.02	5.45 ± 0.38
APR1b	-5.79 ± 0.03	4.48 ± 0.04	0.45 ± 0.01	4.26 ± 0.15
APR2	-5.54 ± 0.08	3.33 ± 0.08	0.44 ± 0.04	2.17 ± 0.32
APR3	-5.50 ± 0.01	2.46 ± 0.07	0.76 ± 0.03	2.79 ± 0.12
APR4	-3.90 ± 0.01	1.71 ± 0.07	0.79 ± 0.04	2.28 ± 0.11

 R_H is, strictly speaking, only true for metals with a spherical surface. For amorphous and i-alloys containing transition metals, due to hybridization of s, p, and d bands, the measured R_H -values were found to deviate greatly from the free-electron value [11, 5]. But for our samples, with a predominantly single type of carrier, the variation of the n_{eff} obtained with temperature (as seen in figure 6) should qualitatively retain the intrinsic behaviour of the sample. n_{eff} is seen to be in the range $10^{19}-10^{20}$ cm⁻³ between 30 and 300 K. μ_H almost keeps at a constant value ~1.9 cm² V⁻¹ s⁻¹ in the temperature ranges 70–300 K for APR1a and 120–300 K for APR1b. A constant value of μ_H is difficult to understand here. Nevertheless, it does indicate that $\sigma \propto 1/R_H$ —that is, σ is proportional to n_{eff} in these temperature ranges. The fact that, above 170 K, σ for APR1b is higher than that for APR1a is therefore due to the former sample having a higher value of n_{eff} . However, the main reason that below about 120 K the σ for APR1b decreases faster than that for APR1a is the rapid reduction in the mobility of the carrier rather than the decrease in n_{eff} for the APR1b sample.

Now a question may be raised: if neither conventional VRH nor WL and EEI theories applied to disordered materials can explain the low-temperature $\sigma(T)$ of Al–Pd–Re QC, what possible mechanism can cause the increase of $\sigma(T)$ for Al–Pd–Re QCs as the temperature is increased?



Figure 7. The magnetic susceptibility χ_M as a function of temperature for samples APR1a, APR1b, APR2, APR3, and APR4. A logarithmic plot of χ_C against *T* for these samples is shown in the inset, where χ_C is proportional to $T^{-\alpha}$. Solid lines indicate theoretical fits.

Fujiwara *et al* [26] argue that for Al–Cu–Fe quasicrystals and their crystalline approximants with large unit cells, the gap between two bands in reciprocal space is often very small, and therefore interband transitions due to electron–phonon or electron–electron interaction are possible even at low temperature. Thus, they suggest that the increase of σ with temperature observed in QCs should be predominantly due to interband transitions. A similar idea was also proposed by Mayou *et al* [25]. Although the interband transition (or, equivalently, the mechanism of local hopping between states separated infinitesimally in energy) is quite attractive for interpreting the temperature dependence of σ and even n_{eff} , we think that more rigorous theoretical calculations are still necessary to assess the importance of the role that it plays in the conductivity behaviour of Al–Pd–Re QC at elevated temperature.

The magnetic susceptibilities χ_M as functions of temperature for APR1a, APR1b, APR2, APR3, and APR4 are presented in figure 7. The common features of χ_M are: (1) the value of χ_M is negative, indicating that the main component of all these samples is diamagnetic; and (2) the magnitude of χ_M decreases rapidly with increasing temperature at low temperatures, reaches a minimum value and then increases with increasing temperature. A large variation of the magnetic susceptibility with temperature such as that seen in figure 7 is quite unusual. Although the electron–electron interaction theory for a diamagnet predicts that the magnetic susceptibility of a disordered diamagnet at low temperatures varies as $T^{1/2}$ [8], our plot of χ_M varying as $T^{1/2}$ between 2 K and 50 K does not follow the prediction. Thus, we believe that the anomalous temperature dependence of χ_M observed for Al–Pd–Re QCs is mainly due to the contribution of paramagnetism—that is, χ_M should consist of paramagnetic and diamagnetic components. This can be seen more clearly from figure 8, where the magnetization *M* is plotted against the magnetic field *H* at 5 K, 77 K, and



Figure 8. The magnetization versus magnetic field at 5 K, 77 K, and 300 K for APR1b. Inset: M versus H between 0 and 5 kOe.

300 K for APR1b. From the figure, we can see that at low field, the value of M is positive and responds quickly to an applied field, a characteristic of paramagnetism; at high field, the value of M is negative, indicating that the diamagnetic contribution surpasses the paramagnetic contribution. In addition, we can also see from the low-field region that the magnetization associated with the paramagnetic component drops rapidly with rising temperature. Therefore, the experimental curves of χ_M (see figure 7) versus T will be fitted by the following equation:

$$\chi_M(T) = \chi_0 + aT^{-\alpha} + bT^2$$

where χ_0 is a temperature-independent susceptibility and is negative; the parameters *a*, *b*, and α are found to be positive values. The values determined for χ_0 , *a*, *b*, and α are listed in table 2. The value of χ_0 is seen to vary in magnitude from the smallest value 3.9×10^{-7} emu g⁻¹ for APR4 to the largest value 5.84×10^{-7} emu g⁻¹ for APR1a.

 χ_0 may include the following three major contributions: the diamagnetic susceptibility of ion core electrons χ_{ion} , the Pauli-spin paramagnetic susceptibility χ_P , and the Landau– Peierls diamagnetic susceptibility χ_{LP} of the conduction electrons. χ_{ion} is estimated to be in the range -2×10^{-7} to -5.33×10^{-7} emu g⁻¹, depending on the values of χ_{ion} for each atom in the Al–Pd–Re formula used [27, 28].

It is seen that most of the values determined for χ_0 , as seen in table 2, fall in the range of the estimated values. Therefore, it is hard to estimate the magnitude of the contribution of the χ_{LP} -term unless we can determine the χ_P -term separately. The bT^2 -term has been found in the magnetic susceptibility of Al–Cu–Fe [29] and Al–Pd–Mn [30] QCs and is ascribed to the temperature dependence of the Pauli-spin paramagnetic susceptibility χ_P . The value of the coefficient *b* of the T^2 -term is determined to be in the range 2.17–5.45 emu g⁻¹ K⁻² (see table 2) and is positive for the Al–Pd–Re QCs studied. This is regarded as further evidence for a pseudogap at E_F in QCs [31].

The Curie-like term $\chi_C = aT^{-\alpha}$ with $\alpha < 1$ must originate from the interactions of localized moments existing in the samples. One may argue that this term arises from paramagnetic impurities. However, from table 2 and figure 7, we find that the resistivity ρ at 4.2 K seems to correlate with the value of χ_C . For example, we can see that a sample with higher $\rho(4.2 \text{ K})$ has larger values of χ_C —except the APR1b sample with an additional low-*T* annealing. This is contrary to what is observed for the Al–Cu–Fe series of QCs, where the magnetic susceptibility χ_M is usually positive for the poor-quality sample with lower $\rho(4.2 \text{ K})$, and χ_M is negative only for good-quality samples with higher $\rho(4.2 \text{ K})$ [16, 29].

Therefore, the indication of the correlation between $\rho(4.2 \text{ K})$ and χ_C for Al–Pd–Re QCs may suggest that the observed anomalous magnetic behaviour is an intrinsic property—rather than being due to paramagnetic impurities—of this series of QCs. As for APR1b, its larger value of $\rho(4.2 \text{ K})$, as we have mentioned in previous sections, is caused by additional low-*T* annealing. This low-*T* annealing may induce the change in structure microscopically, due to the rearrangement of atoms, and therefore possibly also leads to the variation of χ_C . Further investigations by means of microscopical experimental tools are required to clarify this point.

The magnetic properties of Al–Pd–Re QCs are quite similar to those of phosphorusdoped Si (Si:P) semiconductor [32]. In Si:P, the $\chi_C = aT^{-\alpha}$ term occurs in the samples with carriers near the metal–insulator transition (MIT) either from the metallic or the insulating side, and is attributed to the behaviour of an assembly of localized spins (associated with P) distributed randomly in space and interacting antiferromagnetically with each other via short-range direct Heisenberg exchange.

The $aT^{-\alpha}$ -behaviour exhibited by Al-Pd-Re QCs may be explained by the same theory because of the following: (1) it also occurs near the MIT; (2) the exchange interaction between localized spins (possibly associated with Re atoms here) should also be antiferromagnetic as is evident for many Mn-containing QCs; and (3) the transition metal atom, though not distributed randomly, has a distribution of sites as revealed in Mössbauer studies [33], which should also give rise to the broad distribution of exchanging energy necessary for the appearance of $aT^{-\alpha}$ -behaviour. The above arguments, of course, are only tentative. Further theoretical and experimental studies are still needed to ascertain the origin of the $aT^{-\alpha}$ -behaviour observed in the magnetic susceptibility of Al-Pd-Re QCs.

4. Summary

(1) The low-temperature conductivity behaviour of Al–Pd–Re QCs cannot be explained by the theories of WL and EEI, indicating that the conduction electrons in these series of QCs are localized more strongly then those in other QCs.

(2) The temperature-dependent conductivity of Al-Pd-Re QCs can be described by a power law, that is, $\Delta\sigma(T) \propto T^{\alpha}$. It is found that α is roughly equal to 1 for poor-quality samples with higher conductivity, while α is different in both the low- and high-temperature regimes for higher-quality samples with lower conductivity.

(3) At high temperatures, the conductivity of Al–Pd–Re QCs is found to correlate with the inverse of the Hall coefficient.

(4) The magnetic susceptibility χ_M of Al-Pd-Re QCs is negative, and its magnitude is seen to be proportional to $T^{-\alpha}$ at low temperatures, but proportional to T^2 at high temperatures.

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