PHYSICAL REVIEW B VOLUME 55, NUMBER 18 1 MAY 1997-II

Evidence for finite conductivity of icosahedral AlPdRe at $T=0$ K

M. Ahlgren
Fasta Tillståndets Fysik, Kungliga Tekniska Högskolan, S-100 44 Stockholm, Sweden

C. Gignoux
Laboratoire d'Etudes des Propriétés Electronique, CNRS, Boîte Postale 166, 38 042 Grenoble, Cedex 9, France

M. Rodmar
Fasta Tillståndets Fysik, Kungliga Tekniska Högskolan, S-100 44 Stockholm, Sweden

C. Berger
Laboratoire d'Etudes des Propriétés Electronique, CNRS, Boîte Postale 166, 38 042 Grenoble, Cedex 9, France

Ö. Rapp
Fasta Tillståndets Fysik, Kungliga Tekniska Högskolan, S-100 44 Stockholm, Sweden

(Received 26 December 1996)

Samples of icosahedral AlPdRe have been studied for a range of varying resistance ratios $R=\rho(4.2\text{ K})/\rho(295\text{ K})$. Structural characterization techniques used include x-ray diffraction, scanning electron microscopy, and diffraction in transmission electron microscopy. The electrical resistance was measured down to 40 mK. The conductivity at $T=0$ K, $\sigma(0)$, extrapolated from these results was nonzero for all samples and $\sigma(0)$ was found to decrease exponentially with increasing R over more than an order of magnitude in $\sigma(0)$. [S0163-1829(97)51418-4]

The question whether high-resistivity icosahedral quasicrystals are metallic or not has been strongly debated in recent years. Soon after the discovery of stable i-AlPdRe,[1] it became clear that previously known anomalous transport properties of quasicrystals often were even more anomalous. In fact, several observations might suggest that high-resistivity i-AlPdRe is on the insulating side of a metal-insulator (MI) transition:[2] (i) the high resistivity itself,[3–7] (ii) the strong temperature dependence of the resistivity, as measured, e.g., by the resistance ratio $R=\rho(4.2\text{ K})/\rho(295\text{ K})$,[6,7] (iii) the transition, with increasing R values from a giant positive magnetoresistance[8] to small values of variable sign,[5,9,10] and of magnitude observed in some known metal-insulator transition systems, (iv) extrapolations of the conductivity on linear T scale from temperatures above 0.4 K to zero conductivity at finite temperature,[6,10] and most recently, (v) the observation that the temperature-dependent part of the conductivity can be described by variable range hopping expressions for pure and Mn-doped i-AlPdRe.[11]

On the other hand, the low-temperature specific heat is metallic with a low density of states also for samples with large values of R.[6,12] There is no gap observed in high-sensitivity photoemission experiments but instead, again, a reduced density of states.[13] Recent tunnel spectroscopy results display a similar single and narrow pseudogap.[14] Furthermore, with the exception of (v) above, where impurities were assumed *ad hoc* to account for a nonzero conductivity at zero temperature, it has not been possible to account for the low-temperature conductivity, either by activation across a gap, or from established theories for nearest-neighbor or variable range hopping.

In this paper we study the low-temperature conductivity of high-quality i-AlPdRe with the aim to determine whether the zero-temperature conductivity is zero or not. The traditional way to investigate such a problem is to extrapolate the conductivity, $\sigma(T)$, to $T=0$ K and decide if $\sigma(0)$ is zero or nonzero. The main problem to address for i-AlPdRe is then to control sample stoichiometry and any influence of possible impurities in the samples.

In i-AlPdRe a range of R values can be obtained for different samples, all of which are of high quality in the sense that x-ray diffraction shows only a single-phase icosahedral structure. A possible reason for these varying R values may be strongly varying properties of the icosahedral phase, e.g., due to small variations of the Re content in a solubility range.[15] i-AlPdRe samples offer an interesting possibility to handle this problem, since the controlled variation of R values to some extent resembles varying doping concentration in traditional MI systems. If there is no solubility range in the i-phase of AlPdRe, the varying R values must be due to impurities. We investigated if precipitates of a second phase, at a level consistent with the structural investigations, could give an erroneous interpretation of the results.

In all cases we found that the conductivity of i-AlPdRe remains >0. Furthermore a consistent trend was found for the extrapolated values of $\sigma(T=0)$ with increasing R, which leads us to conjecture that this conclusion remains valid also for higher R values than those studied up to now.

Ingots of nominal composition $Al_{70.5}Pd_{21}Re_{8.5}$ were prepared in an arc furnace in purified argon atmosphere. The samples were then remelted in an electromagnetic levitation oven to ensure homogeneity. Ribbons typically 30 μm thick, were prepared by melt spinning and subsequent annealing.

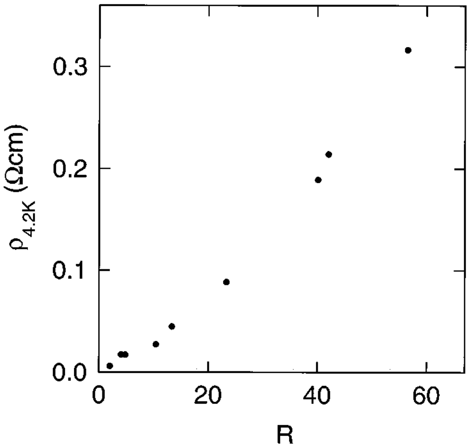

FIG. 1. The resistivity at 4.2 K vs residual resistance ratio $R = \rho(4.2\ \text{K})/\rho(295\ \text{K})$ for several AlPdRe samples of the icosahedral phase.

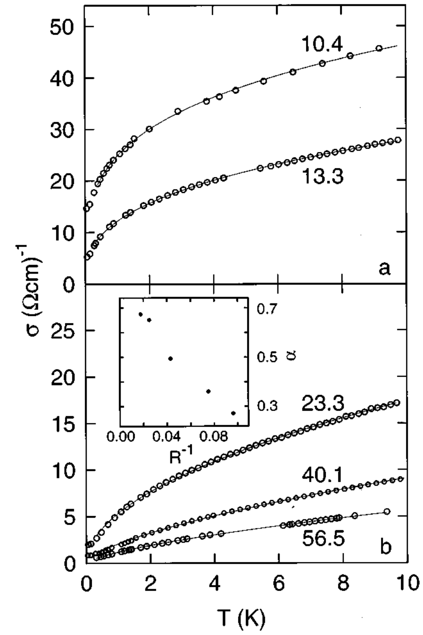

FIG. 2. The conductivity below 10 K for *i*-AlPdRe. The R value for each sample is given in the figure. The full curves are expressions of the form $\sigma(T) \approx \sigma_1 T^{\alpha}$ fitted to each curve. Inset: α of these fits vs R^{-1}.

Different annealing temperatures in the range 800–1020 °C were used to improve sample homogeneity. The total annealing time was usually about 20 h, including furnace cooling to room temperature. The more resistive samples were obtained from annealing at the higher temperatures.[15] Standard x-ray diffraction techniques showed that the samples were of the icosahedral phase. Electron diffraction on selected samples from the same batches confirmed the icosahedral structure.[15]

The resistivity of the samples was measured with a standard four-probe technique in a dilution refrigerator below 1.5 K and in a He^4 cryostat at higher temperatures. Resistivity measurements on small samples of irregular surface shapes are quite inaccurate. However, it can be seen in Fig. 1 that ρ at 4.2 K is closely linear in R. Accordingly we can use R as a convenient parameter to characterize *i*-AlPdRe samples. It is interesting to note that the relation in Fig. 1 is different from that obtained previously[16] for icosahedral AlCuFe, AlPdMn, and AlCuRu, where it was found that σ(4.2 K) was linear in σ(300 K).

Detailed x-ray diffraction studies were made in the region of one of the major diffraction peaks of the icosahedral phase, (230012), for samples with different R values in the range 2–60.[15] Small shifts in this peak with varying R could be observed, corresponding to shifts in $d = \lambda/2\ \sin\theta$ of 0.1% or below. This result indicates some (small) solubility range of the icosahedral phase.

For the low-temperature measurements the thermometer and the samples were attached to a copper block of high thermal conductivity to ensure good thermal contact. Two different sets of measuring equipment were used. The resistance of samples with $R < 23$ was measured with a Guildline current comparator with a minimum current of 1 μA. For more resistive samples, this current could produce self-heating of the samples. We then used a Keithly current source, with currents in the range 10–100 nA and a picovoltmeter as voltage sensor.

The conductivity of five samples with R in the range 10.4–56.5 is presented in Fig. 2 for temperatures up to 10 K. In this temperature range a simple power law $\sigma(T) \approx \sigma_1 T^{\alpha}$ describes data well as illustrated by the curves in the figure. However α does not assume a constant value, e.g., $\frac{1}{2}$ as often observed in barely metallic doped semiconductors.[17] In contrast, α varies gradually with R, and $\sigma(T)$ becomes more linear with increasing R, as observed previously by Poon and co-workers.[6] However, the closely linear relation between α and R^{-1} shown in the inset of Fig. 2 suggests that a strictly linear $\sigma(T)$ is not obtained in this temperature range for any R.

We also investigated an alternative expression, which sometimes has been used,[17] of the form $\sigma(T) = \sigma(0) + \beta\sqrt{T} + \gamma T$, where the linear T term would arise from weak localization effects. Although good descriptions of $\sigma(T)$ could be obtained by this function, the variable signs obtained for γ, and the trend for β would seem unphysical. Fitting this expression to the smoothly varying $\sigma(T)$ of *i*-AlPdRe therefore appears to be overflexible and this attempt was discarded.

The relation shown in Fig. 2 cannot be used for reliable extrapolations to $T = 0$ since there is a change of curvature in $\sigma(T)$ at the lower-temperature end, barely visible on the scale of Fig. 2. Results in the temperature region 40–300 mK are shown in Fig. 3. It is seen that $\sigma(T)$ for all samples approach finite values as $T \rightarrow 0$.

Values of $\sigma(0)$ were estimated from these data by fitting expressions of the type $\sigma(T) = \sigma(0) + \sigma_2 T^x$. Good descriptions of the results were obtained, as illustrated by the curves in Fig. 3, which provides for a useful way to extrapolate data to $T = 0$ K. However, in this case x was found to vary between 1.3 and 2.6 in a way which did not depend systematically on R.

Figure 4 illustrates a remarkably simple apparent linear relation between the logarithm of the extrapolated values of $\sigma(0)$ and $1/R$. The full circle is a sample with $R = 94$ measured with different thermometers and instruments.[18] It is seen to follow the trend described by the other samples. The results suggest that $\sigma(0) > 0$ also for samples of larger R. In fact, the lower bound of $\sigma(0)$ for any of our samples of *i*-AlPdRe would appear to be 0.25 $(\Omega\ \text{cm})^{-1}$, considerably larger than a previous estimate[6] of 0.01 $(\Omega\ \text{cm})^{-1}$. However

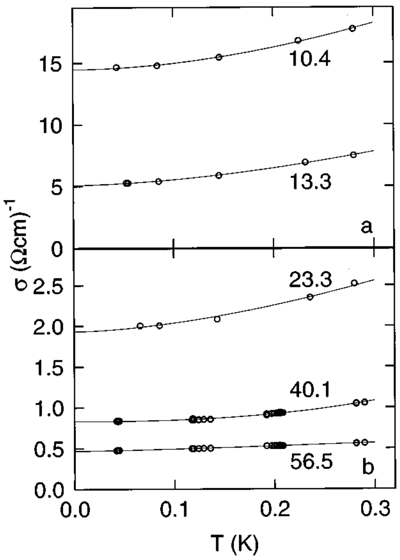

FIG. 3. The conductivity in the range 40–300 mK for the same samples as in Fig. 2. The curves are fits used to extrapolate $\sigma(T)$ to $T=0$ K.

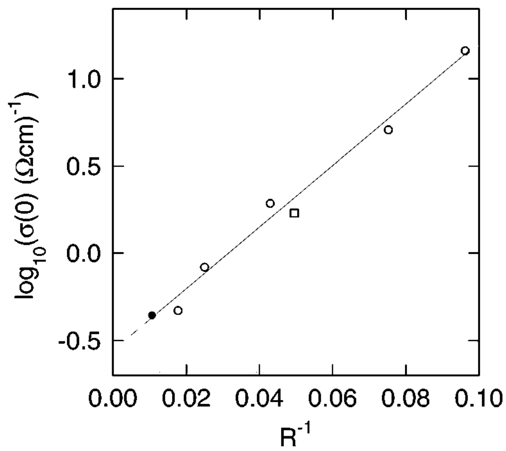

FIG. 4. Logarithm of the extrapolated $\sigma(0)$ vs R^{-1}. The filled circle was obtained from a separate measurement (Ref. 18). For the square, data for $\sigma(0)$ and α were given in Ref. 22, giving a corresponding R^{-1} from the inset of Fig. 2. The straight line is a guide to the eye suggesting a simple exponential dependence of $\sigma(0)$ on R^{-1}.

that estimate was obtained from measurements above 0.45 K.

We discuss various circumstances which might raise objections to these conclusions. First, loss of thermal contact between sample and sample holder is a notorious problem in low-temperature measurements. For the high-resistivity samples we measured the resistance at low temperatures for a series of different currents in the range 10–100 nA, thus varying power dissipation by two orders of magnitude. The sample resistance was the same, as illustrated in Fig. 3 by the overlapping data points for three samples at the lowest temperatures.

Another point of concern is the possibility of thermal decoupling of the electrons at low temperatures. This would happen if, e.g., the phonon scattering relaxation time becomes long enough that the electron gas cannot cool over the finite length of a sample. Observations in two-dimensional Si metal-oxide field effect transistors have been interpreted along these lines.[19] We do not believe that this mechanism is relevant in our case. Saturation due to this mechanism would occur at higher temperatures with increasing mobility. For our samples one expects a decreasing mobility with increasing ρ, but the trend of the saturation region in Fig. 3 is opposite; saturation seems to occur at higher temperatures for the most resistive samples, as is also confirmed for the sample with $R=94$.[2]

Questions about the reason for varying R values in icosahedral samples must also be addressed. A small solubility range in the icosahedral phase was indicated by the x-ray diffraction results as mentioned above, and varying R values would thus, at least to some extent, be an intrinsic property. Can the residual conductivity be due to impurities? Investigations in a scanning electron microscope (SEM),[15] in fact showed that a small concentration of precipitates of size of order 1 μm could be detected in several samples. These spherical-like particles are presumably rich in Re. No apparent correlation was found with the R value of the samples.

The effective conductivity of two-phase composites has been studied in various models. Spherical particles of conductivity σ_B and concentration f_B in an insulating phase can give a resulting conductivity $\sigma^*>0$ only above a percolation threshold of $f_B=\frac{1}{3}$ or 0.25 in different calculations.[20] More complex geometrical forms considered by Yoshida[21] gave similar results. In our samples no impurities were observed in standard x-ray diffraction, suggesting an upper limit for f_B of 0.1, which is considerably smaller than these estimates.

One must also ask if a conducting phase could develop a percolating network, e.g., in grain boundaries, and at a small concentration escaping detection in our various analyses. If so, the measured resistivity would reflect that of the conducting phase. However, for the sample with $R=10.4$, quantum interference effects (QIE's) still account quantitatively for the magnetoresistance. In fact, when the resistivity of this sample was used as a fitting parameter in the analysis of QIE's, a result for ρ was obtained which agreed, within experimental accuracy, with the measured value.[8] This fact assures that the scale of the conductivity is due to the icosahedral phase for this sample, and makes it unlikely that this would change for samples with larger R values, where the concentration of a second phase is expected to be smaller. We conclude that impurities cannot account for the observations of $\sigma(0\ \mathrm{K})>0$.

In addition, our analysis is supported by the consistent relations obtained for R^{-1} vs α in the inset of Fig. 2 as well as for $\sigma(0)$ vs R^{-1} in Fig. 4. In fact these relations appear to provide for a useful way to empirically characterize different i-AlPdRe samples. An example is given by the resistance measurements of Ref. 22, where a sample of nominal composition $Al_{70}Pd_{21.4}Re_{8.6}$ was measured to low temperatures. This is close to but not identical to our nominal composition. R was not given in Ref. 22, but $\sigma(0)$ and an exponent α in a temperature range above the saturation region were extracted. Using the result of $\alpha=0.5$ from Ref. 22 in the inset of Fig. 2 suggests $R\approx 20$, and the corresponding coordinates, shown by the square in Fig. 4, are seen to fit well into our relation between $\sigma(0)$ and R^{-1}.

Summarizing, we have studied a series of high-quality

i-AlPdRe samples of varying resistance ratios. By extrapolating the measured conductivity below 300 mK to $T=0$ K, a nonzero conductivity was found in all our samples. This zero-temperature conductivity decreases exponentially with increasing R, in a way that suggests a finite value of $\sigma(0)$ for all values of R.

We are grateful to G. Fourcaudot and J. C. Grieco for help in the sample preparation and to C. Paulsen for the low-temperature measurements of one sample. Financial support is gratefully acknowledged from EC, Contract No. ERBCHRX-940528, The Region Rhônes Alpes, and the Swedish Natural Science Research Council.

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