Evidence for an insulating ground state in high-resistivity icosahedral AlPdRe from the magnetoresistance

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Magnetoresistance (MR) has been studied between 1.5–8 K in magnetic fields B up to 12 T in icosahedral (i)-AlPdRe samples prepared by two different methods and with resistance ratios $R[=\rho(4\,\mathrm{K})/\rho(295\,\mathrm{K})]$ from 45–160. The observed temperature- and field-dependence of the MR could be well described by variable-range hopping of the Efros-Shklovskii type. The results give compelling empirical evidence for an insulating ground state of i-AlPdRe for $R\geqslant 45$, and suggest that the metal-insulator transition in i-AlPdRe can be monitored by the R value.

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The possibility of a metal-insulator transition (MIT) in highly resistive quasicrystals is a current issue of great challenges and controversies. Strongly different results, theoretical as well as experimental, continue to appear. For instance, the contrasting views have been advanced that a clean quasicrystal at $T=0$ K should[1] or may[2] be insulating, or that it should have a finite conductivity.[3] Recent experimental results for the conductivity $\sigma(T)$ of icosahedral (i)-AlPdRe at low temperatures are also remarkably different.[4–8] For example, evidence has been obtained for an insulating ground state from the observation of Mott variable-range hopping (VRH),[4] while the results for differently prepared samples of the same nominal composition could not be described in this way unless a finite zero-temperature conductivity $\sigma(0)$ was assumed.[5,6] There is no agreement on the interpretation of $\sigma(T)$. Published results[4–8] include propositions of both Mott and Efros-Shklovskii (ES) VRH, as well as estimates for the characteristic temperatures T_0 of the activated hopping that cover a range of six orders of magnitude with ensuing differences in purported localization lengths. It thus appears that results for $\sigma(T)$ cannot clarify whether high-resistivity i-AlPdRe is insulating or not, nor the nature of low-temperature transport.

These difficulties may be due to several problems. The phase diagram of i-AlPdRe and relevant microstructural details are not known, and one cannot at present distinguish, e.g., if a finite $\sigma(0)$ is an intrinsic icosahedral property or if it is associated with quasicrystalline defects or with minute precipitates of a metallic phase in the grain boundaries. In addition, measurements at the lowest possible temperatures are desirable, but the low thermal conductivity of i-AlPdRe make such measurements increasingly difficult.

The aim of the present paper is to explore the alternative to study localization behavior of i-AlPdRe by the magnetoresistance (MR). This is a powerful technique since it circumvents problems with possible impurity phases. Contributions to the MR from a high-resistivity impurity phase are unlikely, as shown previously.[9] Possible minor metallic impurity phases, perhaps accounting for a finite $\sigma(0)$, would have a resistivity that is smaller by several orders of magnitude compared to the quasicrystalline phase, and the corresponding MR is then negligible.[10]

We have studied the MR of four samples of i-AlPdRe with two samples from each of the two groups mentioned above with strongly different low-temperature conductivities.[4–6] The resistivity ratios R $[=\rho(4\,\mathrm{K})/\rho(295\,\mathrm{K})]$ were in the range 45–160, i.e., beyond the weak-localization region, and it has not previously been possible to describe the MR of such quasicrystals quantitatively. The measurements were focused on the region around the minima in the MR for the present samples from 1.5–8 K in fields up to 12 T, and the unresolved problems concerning the low-temperature conductivity were thus avoided. A consistent description is obtained for all samples, giving compelling evidence for ES VRH and, thus, the fact that an insulating ground state is an intrinsic property of i-AlPdRe at $R\geqslant 45$. Evidence is presented that the readily accessible parameter R can be used to monitor the MIT.

All samples were of the nominal composition $Al_{70.5}Pd_{21}Re_{8.5}$. We studied two samples prepared by method A with melting in an arc furnace and subsequent annealings followed by quenching, and two by method B with melting in an arc furnace followed by melt spinning and annealings and terminating with furnace cooling. Samples from method A had typical dimensions of $1\times 1\times 5\,\mathrm{mm}^3$ while samples from method B were foils of thicknesses of about 30 μm. The samples were of a pure icosahedral phase in standard x-ray diffraction (XRD). Some details of the preparation and characterization techniques were used previously and included XRD and scanning electron microscopy.[9] Measurements were made up to 12 T in a flowing gas cryostat using four-probe silver contacts and dc techniques. The temperature drift during field sweeps was reduced to a few mK, using a high-purity copper sample holder extending into a field-free region above the magnet. Temperature errors are therefore estimated to be negligible.

Measurements of the MR, $\Delta\rho(B,T)/\rho(0,T)$, were made on samples with $R=45$ (method B) and $R=160$ (method A), and were analyzed together with recent measurements of the MR for two samples with $R=98$ (method B) and $R=107$ (method A).[9] The observed MR is shown in Fig. 1 for the $R=160$ sample. Similar results were found for the other samples as illustrated by graphs for the $R=98$ and 107

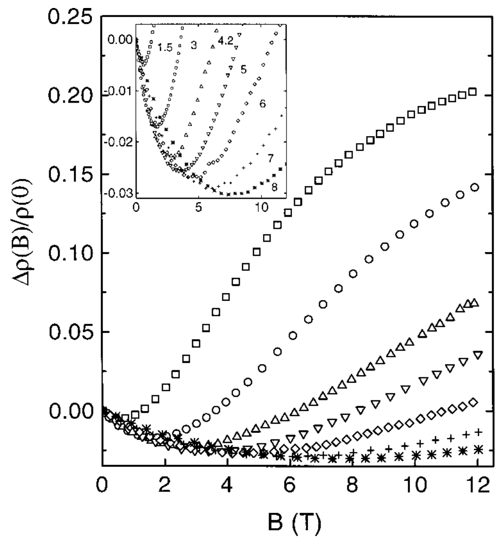

FIG. 1. The magnetoresistance vs B for the $R=160$ sample of i-AlPdRe between 1.5–8 K. Inset: enlargement in the region of the minima, with temperatures in kelvin given on the curves. The sequence of temperatures is the same in the main panel.

samples shown previously.[9] Some common features are the following: (i) The MR is negative and linear in B for small fields, passes through a minimum at a field $B_{\min}$ and increases with further increasing B. (ii) At larger magnetic fields the MR increases more slowly. (iii) With decreasing temperature, $B_{\min}$ and $|\mathrm{MR}(B_{\min})|$ decrease. Similar behavior was observed in earlier work on quasicrystals.[7,9,11] In other cases, e.g., lightly doped semiconductors, the negative MR can decrease[12,13] or increase[14,15] with decreasing temperature depending on the system.

The general features of our results are similar to numerical simulations on hopping conduction.[16] The work by Shklovskii and Spivak[17] was therefore taken as a starting point for the analyses. In this model interference of the contributions to the amplitude of the hopping probability from different trajectories gives a negative MR,

$$\ln\frac{\rho(B)}{\rho(0)}\approx\frac{\Delta\rho(B)}{\rho(0)}=-\frac{k}{B_0}B, \tag{1}$$

while a positive contribution arises from the shrinking of wave functions in magnetic field and in the limit of weak fields, with $\Delta\rho(B,T)/\rho(0,T)\ll 1$. It increases as B^2;

$$\ln\frac{\rho(B)}{\rho(0)}=\frac{1}{B_0^2}B^2. \tag{2}$$

Eq. (1) was fitted to the low-field region below $B_{\min}$ and Eq. (2) to the data above $B_{\min}$. This method uses two straightforward analyses, each with one adjustable parameter, and therefore best suited to determine k and B_0.[18] The inset of Fig. 2 illustrates these procedures on a linear B scale for the $R=160$ sample at 4.2 K. Similar fits in the linear negative and parabolic positive regions are characteristic for all data.

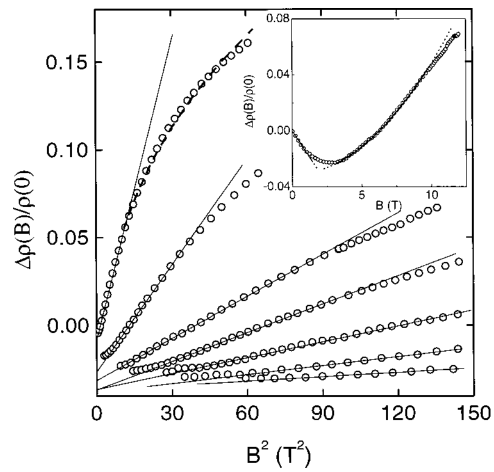

FIG. 2. The MR vs B^2 above the minima for the $R=160$ sample. Temperatures are in the same sequence as in Fig. 1. The dashed line above 4 T at 1.5 K has the form $\Delta B/B\sim B^{2/3}$ and is discussed in the text. Inset: fits of Eqs. (1) and (2) for the same sample at 4.2 K.

The main panel in Fig. 2 shows the MR of the $R=160$ sample vs B^2. Over a range of fields that increases with increasing temperature, Eq. (2) describes the data well. At larger fields there are deviations in the direction of a smaller MR. The dashed line at 1.5 K in Fig. 2 illustrates that the B^2 region (solid line) is followed by a $B^{2/3}$ behavior up to about 7 T in qualitative agreement with the Shklovskii-Efros theory.[19]

We now analyze the coefficients of Eqs. (1) and (2). B_0 is the characteristic field for which a flux quantum passes through the interference area $(r^3\xi)^{1/2}$, where ξ is the localization length and r the hopping distance.[17,20] For ES VRH, the temperature dependence of r is given by $r=(\xi/4)(T_0/T)^{1/2}$, and the temperature dependence of the coefficient of the positive MR, Eq. (2), is then

$$B_0^{-2}=0.0015\frac{e^2\xi^4}{\hbar^2}\left(\frac{T_0}{T}\right)^{3/2}=\beta T^{-3/2}. \tag{3}$$

T_0 is the characteristic temperature in ES VRH, and the proportionality constant was estimated by Efros and Shklovskii.[19] With $r=(3\xi/8)(T_0'/T)^{1/4}$ for Mott VRH one has $B_0^{-2}(T)\sim T^{-3/4}$, while nearest-neighbor hopping would give a nearly temperature-independent coefficient. $B_0^{-2}(T)$ vs $T^{-3/4}$ and $B_0^{-2}(T)$ vs $T^{-3/2}$ are shown in Fig. 3. The present samples are clearly seen to obey ES VRH.

Considering interference events from all tunneling paths within the approximation of one scattering event only, Raikh and Wessels[20] obtained quantitative predictions for the negative linear MR. The parameter k of Eq. (1) is a function of the normalized scattering amplitude. Varying $k(T)$ behavior has been suggested in the literature. Agrinskaya *et al.* found in a semiphenomenological model that dk/dT is negative for Mott VRH and positive for ES VRH.[21] Experiments with CdTe of varying dopings confirmed this by observations of a negative (positive) dk/dT in the MR in conjunction with Mott (ES) VRH in $\sigma(T)$.[12] We calculated k from B_0 and the

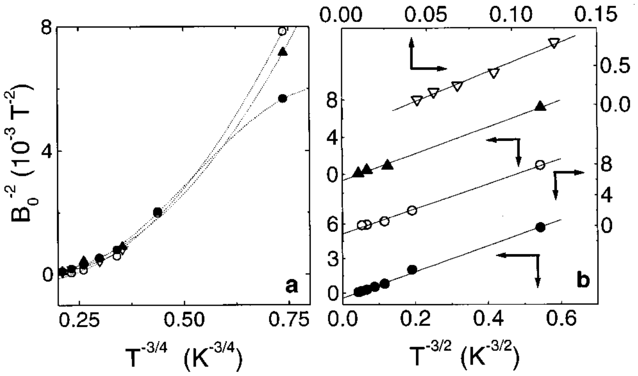

FIG. 3. Temperature dependence of B_0^{-2} for (a) Mott VRH and (b) ES VRH. R values are ▽: 98, ○: 45, ▲: 108, and ●: 160 in both panels. (b) Data were vertically displaced, and scales in the same units as in (a) are indicated by the arrows. ES VRH is obeyed in all cases.

slope of the MR at small B. k was found to increase with increasing T in all samples, approximately[21] as $T^{1/2}$, which is illustrated in the inset of Fig. 4 for one sample. These observations thus also support ES VRH.

Eqs. (1)–(3) and the result for $k(T)$ give $B_{\rm min}(T) \sim kB_0 \sim T^{1/2}T^{3/4} \sim T^{5/4}$. The observed $B_{\rm min}$ for two samples is shown in the main panel in Fig. 4 and is shown to obey this behavior. Similar temperature dependence was obtained for all samples. This result gives further support for the predictions of ES VRH and the consistency of the analyses.

R values of *i*-AlPdRe are roughly linearly correlated with ρ(4 K),[6] which for the present samples ranges from about 250–2100 mΩ cm, when R varies from 45–160. However, as seen in Fig. 3(b), β [of Eq. (3)] does not depend strongly on R. For all samples β is within $1.2\times10^{-2}\pm20\%$ (T^{-2} K$^{-3/2}$). With[20] $k \sim T_0^{-1/2}$, the variation of k due to differences in T_0 between the samples is also small. One thus expects that variations of k among the samples are mainly due to the resistivity. This is confirmed by our results. As shown in Fig. 5, k increases with R, reflecting an increased scattering strength with increasing R. k vs R is similar for A and B samples above 6 K. At lower temperatures k is more uncertain, since the minimum of the MR becomes more shallow (Fig. 1), but the results suggest that data for B samples start to fall below those of A samples. This may be a precursor to the different MR for A and B samples below 1.5 K observed previously.[9] The results in Fig. 5 indicate that the MIT is likely of the Anderson type. Furthermore, an extrapolation to $k=0$ in Fig. 5 gives $R\approx30$ in qualitative agreement with Ref. 9, where the MR>0 at $R=23$ and negative at low fields for $R=45$ at comparable temperatures. Our results indicate that the appearance of a negative MR is associated with a transition to an insulating state, and that R appears to be a convenient parameter to monitor the MIT.

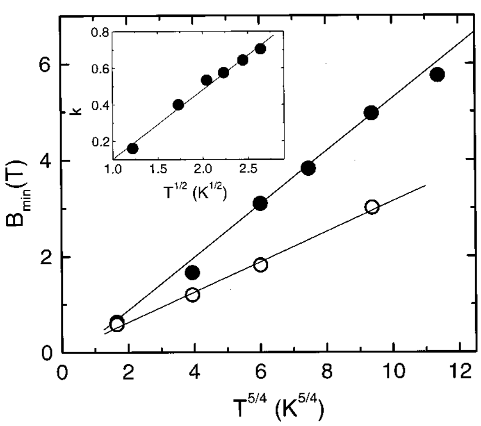

FIG. 4. $B_{\rm min}$ vs $T^{5/4}$ for two samples $R=45$:○ and $R=160$:●. Inset: k vs $T^{1/2}$, characteristic for ES VRH for the $R=160$ sample.

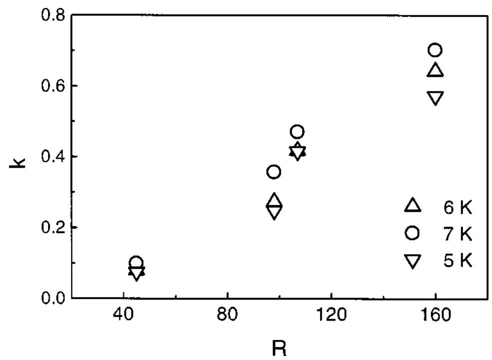

FIG. 5. k of Eq. (1) vs R at the temperatures indicated. k increases with R at all temperatures, reflecting increased electron scattering.

$\sigma(T)$ was analyzed over the temperature range of the MR measurements. For both Mott and ES VRH, a constant $\sigma(T=0\ {\rm K})$ had to be added to the VRH expressions to obtain reasonable descriptions and in both cases similar good fits were obtained. We thus cannot separate different VRH behaviors. In addition to the unresolved nature of $\sigma(0)$, the numerical flexibility with three fitting parameters and an exponential function make quantitative results from $\sigma(T)$ quite uncertain,[22] as mentioned before.

More detailed results from the MR alone would be desirable. However, the MR at $B\leqslant B_{\rm min}$ depends on k and the scattering amplitude, and is thus also related to understanding $\sigma(T)$. Alternatively one could use the result that MR $\sim B^{2/3}$ above the B^2 region, as predicted by Shklovskii[19,23] and confirmed by our analysis, but the proportionality factor is not known, and this result could not be used quantitatively. Using available estimates[22] it may be suggested that we are in a marginal regime of the model where the distance between carriers is not larger than the magnetic length $(\hbar/eB)^{1/2}$, since quasicrystals have comparatively large charge density. Empirical evidence that data obey ES VRH is compelling, however. Quasicrystals are quite different from conventional doped semiconductors, and further studies of their transport properties may be called for. For example, the carriers within r have energies different from the electron that hops, and one can speculate that these carriers can be viewed as sites, maintaining a ''dilute'' limit.

Recent results of tunneling and point spectroscopy studies on *i*-AlPdRe may give independent support for the applicability of the Efros-Shklovskii model to high resistivity *i*-AlPdRe.[24] These authors observed a deep and narrow pseudogap at 2 K for a sample with $R \approx 100$, while this feature was strongly attenuated for an $R=30$ sample, and they tentatively associated this with enhanced Coulomb interaction effects in samples with larger *R*. Our estimate from Fig. 5 that $R \approx 30$ would roughly correspond to the transition between metals and insulators is also qualitatively consistent with these observations.

We briefly discuss the possibility of contributions to the MR from spin effects. When crossing an MIT, enhanced magnetic interactions could be expected, and spin contributions to the MR in the VRH region have been studied by various authors.[25,26] In particular, the results of Ref. 26 indicate a linear and isotropic low-field MR as in our observations and in Refs. 17 and 20. However, this mechanism does not appear to be applicable here. The magnetic susceptibility, is diamagnetic, and at most, weakly dependent on temperature and *R* over the wide range from $R=3-160$.[27] Furthermore, in the model of Ref. 26, the MR saturates at high fields. In contrast, the MR of *i*-AlPdRe (Fig. 1) develops at moderate fields beyond the B^2 region as predicted by the model of Refs. 16 and 19, and for an $R=115$ sample,[28] the MR does not saturate at higher fields but decreases beyond a maximum at about 20 T.

We have studied the magnetoresistance of *i*-AlPdRe in differently prepared samples and analyzed the temperature dependence of three different parameters *k*, $B_{\min}$, and B_0^{-2}. For all samples and parameters the observations agree with Efros-Shklovskii VRH. These results give unequivocal evidence that an insulating ground state is an intrinsic property in *i*-AlPdRe for $R \geqslant 45$. A relation between *k* and *R* is found, which suggests that the transition is disorder driven and can be monitored by the easily measured parameter *R*.

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[18] A fit of Eqs. (1) and (2) with two free parameters can of course also describe the data. This accounts well for the observed $B_{\min}$ but gives larger scatter in the results for *k* and B_0.

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[22] This uncertainty is illustrated by the following calculations. Close to an MIT, $T_0 \sim 1/\xi^3 g$, where g is the density of states. The slopes of the straight lines in Fig. 3(b) are $\beta \sim \xi^4 T_0^{3/2}$. Solving T_0 in the experimentally accessible g and β gives $T_0 \sim \beta^6 g^8$ showing that these relations are extremely ill-conditioned for numerical work. Instead, using T_0 as obtained from $\sigma(T)$ is quite uncertain as illustrated in the text for Mott VRH. If we take $T_0 \sim 12$ K from our results for $\sigma(T)$ between 1.5–7 K, an order-of-magnitude estimate might be $\xi \approx 150$ Å, and $r(1.5\,\mathrm{K}) \approx 100$ Å. The error in these numbers may easily be a factor of 3.

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