Monitoring an insulator-metal transition in icosahedral AlPdRe by neutron irradiation

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The intrinsic disorder effect on the electrical resistivity and magnetoresistance (MR) of icosahedral AlPdRe is studied by use of high-energy neutron irradiation. The icosahedral phase is preserved under irradiation with a decrease of x-ray peak intensity and volume of the coherent icosahedral phase. An insulator-metal transition, as observed in the MR, can be driven by the irradiation and is monitored by the resistance ratio R $[=\rho(4.2\text{ K})/\rho(295\text{ K})]$. The relation of MR vs R was found to be similar for samples of a widely different history.

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Icosahedral (i)-AlPdRe samples can be made with low-temperature resistivities ρ and resistance ratios R $[=\rho(4.2\text{ K})/\rho(295\text{ K})]$ varying over wide ranges of values. Already early studies of high-ρ samples with R of 51 (Ref. 1) or 125 (Ref. 2) suggested insulating properties. It is only recently, however, that more direct evidence of a metal-insulator transition (MIT) in i-AlPdRe has been found. The magnetoresistance (MR) at large R was found[3] to obey Efros-Shklovskii variable-range hopping (VRH) theory for electron transport in insulators.[4] From the metallic side, an MIT is suggested both from analyses of the metallic MR at small R (Ref. 5) and from a scaling approach to the conductivity $\sigma(T)$ of metallic samples at temperatures $T \geqslant 400$ mK.[6] These different investigations all give estimates that an MIT occurs in the region $R \sim 20$–30.

However, the most perspicious results on an MIT are normally expected for low-temperature $\sigma(T)$, but such results for i-AlPdRe have remained controversial. Descriptions in terms of VRH theories have given widely different results.[7–12] A major difficulty is the saturation of $\sigma(T)$ observed at very low temperatures ($\lesssim 20$ mK in high-R samples) and incompatible with VRH. Recent results indicate that a finite $\sigma(0\text{ K})$ is a property of the icosahedral phase,[13] which may suggest that at large R and low T two channels contribute to $\sigma(T)$, e.g., variable-range hopping and quantum tunneling.

Nor has the role of the parameter R been understood. Although it has been provisionally used as a parameter to classify electronic transport results,[3,5,6] justification for this approach has been based on convenience rather than understanding. R is simply easier to measure accurately than ρ. A relation between R and structural properties has not been found for i-AlPdRe. Nor is it known how to monitor an MIT, since resulting resistive properties are not well controlled in existing preparation techniques.

The starting point for the present work is the question if homogenous disorder by neutron irradiation can be used to tune transport properties of i-AlPdRe. In amorphous metals such experiments have been found to increase structural *and* electronic disorder, leading to an increased ρ, decreased superconducting T_c, and a small shift in the direction of the MIT.[14] For quasicrystals it has long been known that increased structural disorder instead decreases R and ρ,[15] and this is expected also for i-AlPdRe. We therefore started with an insulating sample and irradiated it with a series of doses D. Using a single sample focuses on the effect of varying intrinsic disorder. For increasing D the icosahedral phase was preserved, peak intensities and R decreased, and an insulator-metal transition was traversed. The MR vs R across the MIT was found to be independent of sample history. In contrast, e.g., to $\rho(4.2\text{ K})$, R can therefore serve as a parameter to control the MIT.

Samples had nominal composition $Al_{70.5}Pd_{21}Re_{8.5}$ and were prepared either by melting in an arc furnace followed by annealings at 940 and 600–650 °C (ingots) or by arc melting followed by melt spinning and annealings (foil sample). Details of these preparation techniques and structural characterizations have been given previously.[5] Bars of dimensions $\approx 1\times1\times3$ mm^3 were cut from the ingots. Most studies were made on such a sample with $R=67$. In addition an ingot sample with $R=27$ and a foil sample ≈ 40 μm thick were also studied. The latter sample was brittle. It broke after the first dose and was not further used.

Samples were packed tightly in a thin-walled Al container for good thermal contact with cooling water and irradiated with fast neutrons of mean energy 1 MeV and flux 5×10^{13} neutrons/cm^2 s, in the nuclear reactor in Zarechnii. These neutrons penetrated the samples. Sample temperature is estimated to remain below 100 °C, i.e., well below all previous annealing temperatures. ^{185}Re is excited by neutrons to ^{186}Re, which is a strongly radioactive isotope with half-life of 4 days. After $\sim$1 month the radioactivity of the samples was about $(1–100)\times10^{-6}$ rad/s at 5 cm distance, and they could be handled with tweezers. Electrical transport measurements were made by standard four-probe techniques in magnetic fields up to 13.6 T. Contacts of In-Ga were applied by ultrasonic soldering. They were removed before each new irradiation.

X-ray diffraction data were taken in steps of 0.05° in 2θ from 20° to 76°. An ordered icosahedral phase was preserved for all doses D over a range of more than two orders of magnitude. A diffraction pattern is shown in Fig. 1 for the largest D. The number of observable peaks decreased with increasing dose; 16 i peaks were observed at $D=5$

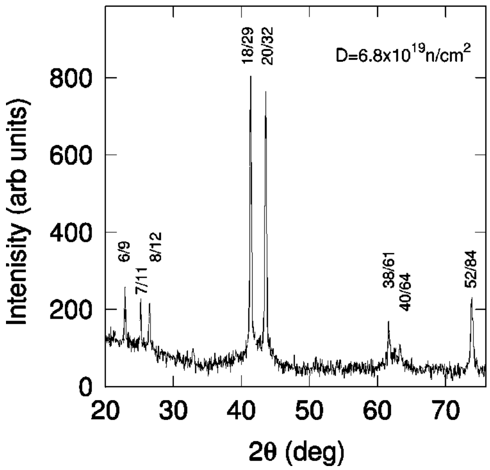

FIG. 1. X-ray diffraction results with Cu $K\alpha$ radiation for the sample with the largest irradiation dose *D*. Eight icosahedral peaks are labeled in the Cahn indexing scheme.

$\times 10^{17}$ neutrons/cm^2 as compared to 8 in Fig. 1, and the decrease of peak height was usually monotonous with *D* as illustrated for five peaks in Fig. 2. One exception is the 38/61 peak, which has a slightly larger peak height at $D=1.8\times 10^{19}$ neutrons/cm^2 than at $D=7.6\times 10^{18}$ neutrons/cm^2. We ascribe such variations to the texture of the bulk sample surface and different surfaces likely being exposed in successive x-ray experiments. No shift in peak position or any systematic change of the widths of the peaks could be observed within our resolution.

There have been only few previous irradiation experiments on stable quasicrystals. With ~ 1 GeV heavy-ion irradiation on i-$Al_{65}Cu_{25}Fe_5V_5$, a decreasing peak intensity was also observed, but in this case the (7,11) peak broadened with initial dose.[16] This is possibly due to strain associated with inhomogenous irradiation damage in the form of long narrow channels. However, from $D=10^{12}$ to 10^{13} ions/cm^2, further peak broadening did not occur and the peak intensity continued to decrease. A similarity to our results can be expected at large doses, since heavy swift ions will then produce a more homogeneous disorder, as for neutron irradiation at ~ 1 MeV.

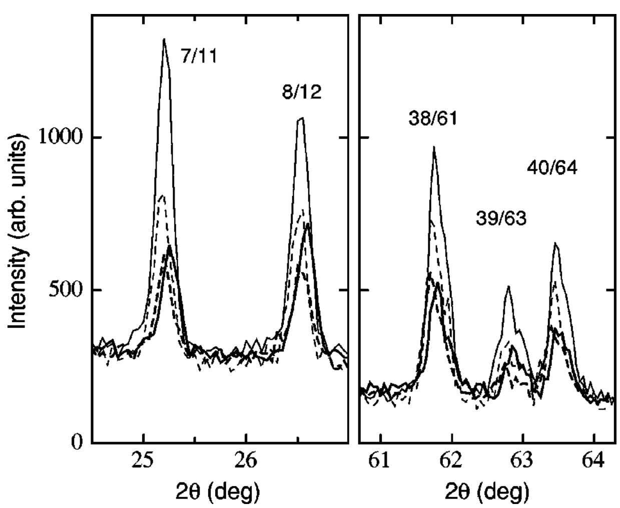

FIG. 2. Five diffraction peaks for varying *D*. Thin solid curve, $D=5\times 10^{17}$; thin dashed curve, 3.6×10^{18}; thick solid curve, 7.2×10^{18}; and thick dashed curve, 1.8×10^{19} neutrons/cm^2. $R_{D=0}=67$.

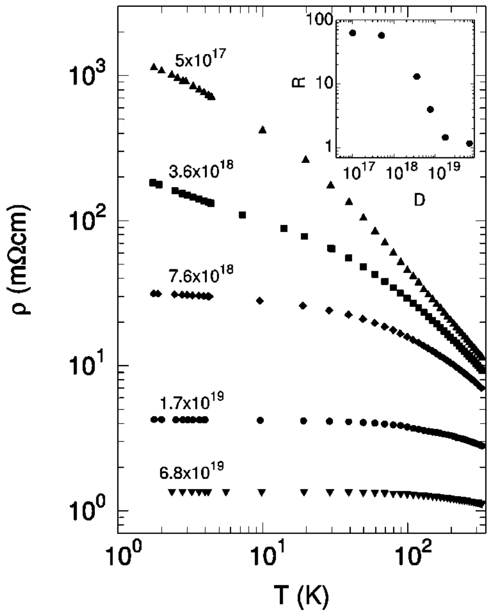

FIG. 3. ρ(T) from 1.5 to 320 K at the irradiation doses (in neutrons/cm^2) given in the figure. $R_{D=0}$ is 67. Inset: *R* vs *D*.

The irradiation thus leads to loss of coherent icosahedral volume. One must ask if these observations are due to second-phase precipitations or reflect a decrease of the coherence length of the long-range icosahedral atomic ordering. Crystalline impurities induced by irradiation can be ruled out since none of the few unindexed peaks, which were occasionally observed for some *D* in the range from 5×10^{17} to 6.8×10^{19} neutrons/cm^2 increased in intensity with *D*. At the largest *D*, unindexed peaks were not observed (Fig. 1). Precipitation of amorphous phases may escape detection by x rays. We therefore made a few annealing experiments on a sample with $D=3.7\times 10^{19}$ neutrons/cm^2. The results indicated reversible behavior. E.g., after 20 min at 350 °C, peak intensities were found to increase. An increase of *R* (by 10%) was also observed, opposite to the trend observed after irradiation. Dissolution of an impurity phase is unprobable at this low temperature. This behavior instead likely reflects a recovery of the icosahedral atomic ordering.

We now turn to the transport properties. ρ and the magnitude of its temperature derivative were found to decrease strongly with increasing *D*, while *R* decreased from 67 in the unirradiated samples to 1.2 at $D=6.8\times 10^{19}$ neutrons/cm^2 (Fig. 3). Two important conclusions can immediately be drawn from these results. First, $\rho(295\text{ K})$ has decreased significantly with irradiation, by a factor ≈ 10 in Fig. 3, in contrast to a set of as-prepared samples with a similar range of *R* values, where $\rho(295\text{ K})$ was close to a constant.[5] Similarly one can find from two samples in Fig. 3 that the ratio $[\rho(4.2\text{ K}, R=13)]/[\rho(4.2\text{ K}, R=4)]$ is 4.4, while the corresponding ratio was 2.6 for two as-made samples with *R*

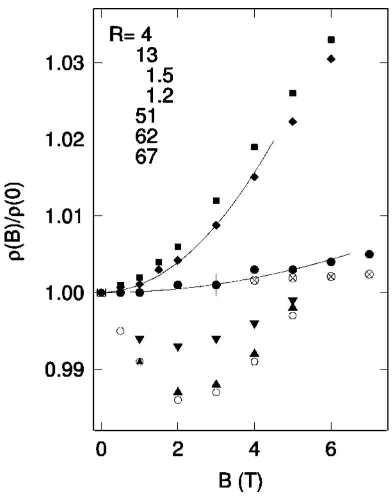

FIG. 4. $\rho(B)/\rho(0)$ vs B at 4.2 K. Here R for the samples varies from top to bottom in the order given in the figure. Typical measurement error is shown for one datum. Curves at $R=13$ and 1.5 are guides to the eye of the form $\Delta\rho(B)/\rho(0)=1+\alpha B^2$. Data below 3 T for the $R=1.2$ sample and above 5 T for the high-R samples have been omitted for clarity.

$=13$ and 4.[5] Obviously the relation between $\rho(4.2\text{ K})$ and R for irradiated and as-prepared samples with similar R values is quite different.

Second, differences in $\rho(4.2\text{ K})$ and R between different samples were found to diminish with increasing D. E.g., at $D=4\times10^{18}$ the sample $R_{D=0}=27$ had $\rho(4.2\text{ K})$ of 63 mΩ cm, still considerably smaller than 130 mΩ cm for the $R_{D=0}=67$ sample at a comparable dose in Fig. 3. However, when these samples were compared at 1.4 and 1.7 $\times10^{19}$ neutrons/cm^2 respectively, $\rho(4.2\text{ K})$ is close to 4 mΩ cm in both cases. In fact, $\rho(T)$ was then almost identical from 2 to 300 K. The structure which causes R to vary in as-made icosahedral samples is thus destroyed beyond a certain defect level. Further irradiation appears to cause similar effects in all samples, irrespective of original states.

The reason for the variation of R in a set of as-prepared samples is not known. Irradiation of a single sample in this respect offers great advantages since several hypothetical reasons can be ruled out. Irradiation-induced phase transformations that influence transport properties are unlikely, as discussed above and further exemplified by the MR results shown below. The impurity content of the starting elements largely dominates any residual remanents of nuclear reactions. Varying chemical composition of the i phase can also likely be ruled out. Nuclear reaction products are negligible. The sample temperature remains below 100 °C, and diffusion is not expected. The experiments instead suggest that the decreasing R and ρ for irradiated samples are related to the introduction of defects in the icosahedral phase and the decreasing coherence length of the ordered i phase.

Figure 4 shows $\rho(B)/\rho(0)$ vs B in the low-field region. For high-R samples (unirradiated $R=67$ and irradiated $R=62$ ingots, and $R=51$ foil sample), the MR at small B is negative, goes through a minimum, and increases to positive values for increasing B. This MR can be well described[3] by

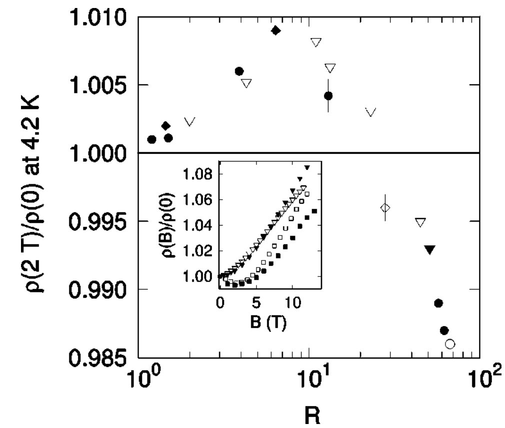

FIG. 5. Magnetoresistance at 4.2 K and 2 T vs R. Open circle (rhomboid): as-prepared samples with $R_{D=0}=67$ (27). Corresponding solid symbols: same samples after irradiations. ∇: as-prepared samples from Ref. 5. Inset: comparison of MR at 4.2 K up to 13 T. Open symbols, as-made samples (Ref. 5) (▽), $R=13.3$, (□) $R=45$, irradiated samples (▼) $R=13$, (■) $R=51$.

Efros-Shklovskii theory for insulators, with a negative MR due to interference between different trajectories contributing to hopping and a positive contribution from shrinking wave functions in a magnetic field.[4] For $R\leqslant13$ in Fig. 4 the MR is positive at all experimental fields. This MR is quantitatively described by weak localization and electron-electron interactions for weakly (electronically) disordered metals.[5] At low fields and not too weak spin-orbit scattering all such contributions increase as B^2, as indicated by curves for two samples. For increasing D, the MR decreases down to 0.1%. At $R=1.2$ the MR and $\rho(T)$ have approached characteristics for amorphous metals where the MR is usually below 0.1% and R rarely exceeds 1.1.[17] A transformation to an amorphous phase can eventually be expected for strong radiation damage. It has been observed in some quasicrystals after ~1 MeV electron irradiation at doses in excess of 10^{26} electrons/cm^2.[18]

A wide range of samples is included in Fig. 4 from well into the insulating side of the transition to far into the metallic side. The results show that neutron irradiation can monitor an insulator-metal transition. This conclusion does not rely on a precisely given location of the MIT. In fact it may not be possible to specify such an R value. MIT is a quantum transition at $T=0$, and in finite-temperature experiments an MIT may appear to occur gradually. The sign change of the low-field MR may give (at least) a rough location of the transition. There is in contrast no obvious feature of $\rho(T)$ in Fig. 3 from which an MIT can be qualitatively identified. This remains valid also for other ways of displaying data, such as $\ln\rho(T)$ vs $T^{-1/2}$ in Efros-Shklovskii VRH theory.

MR studies of icosahedral samples have indicated that impurities do not contribute to observations.[5,19] In particular, irradiation-induced precipitation of an amorphous metal would have small $\rho(4.2\ K)$, compared to the samples in Fig. 3, and a negligible MR.[19] The results in Fig. 5 reinforce this conclusion. At fixed B and T the MR is similar for irradiated as well as non-irradiated samples.

MR vs B at 4.2 K is shown in the inset of Fig. 5 for two sets of differently prepared samples, each set with similar R values. The MR is closely similar up to 10 T for two samples with $R=13$. For the two high-R samples the MR is qualitatively similar, with a somewhat smaller MR at the larger R, as expected from the trend in Fig. 4.

A similar MR for samples of different history and similar R is further illustrated in the main panel of Fig. 5. The MR at 4.2 K and 2 T is shown versus R for three sets of samples, i.e., the present irradiated samples with $R_{D=0}$ of 67 and 27, and unirradiated foil samples from Ref. 5. In all cases the MR follows qualitatively the same relation from negative values at large R's, through a sign change, a maximum, and a decreasing positive MR for a continued decrease of R at small R's. These results suggest that the MR is a unique function of R, with MR(R) independent of the details of intrinsic defects and concentration and the composition of extrinsic or intrinsic impurities.

This conclusion is not valid for MR as function of some parameter from $\rho(T)$ such as $\rho(4.2\text{ K})$. Different defects in the icosahedral phase affect $\rho(4.2\text{ K})$ and R differently but not MR(R). Our results show that R has the more fundamental importance. It is R, the average temperature dependence of $\rho(T)$, rather than ρ itself at a particular temperature which can monitor the MIT.

In summary, three results have emerged from our studies of neutron-irradiated *i*-AlPdRe: Irradiation over a wide range of doses preserves the dominating icosahedral phase, but peak intensity and volume of coherently scattering *i* phase decrease simultaneously with a continous decrease of R. This result gives a first relation for *i*-AlPdRe between structural quality and transport properties. Further, it was found that the MR as a function of R is independent of the detailed nature of impurities or defects. Finally, an insulator-metal transition in *i*-AlPdRe can be produced by neutron irradiation damage and this transition is monitored by the R value. The present findings allow systematic studies of the disorder effect on MIT in the same quasicrystal sample. Thus future investigations will include the nature of the relevant quasicrystalline defects.

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