Low-temperature resistivity and magnetoresistance of the quasicrystalline icosahedral $Al_{80}Mn_{20}$ and decagonal $Al_{78}Mn_{22}$ alloys

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Resistivity and magnetoresistance measurements are reported for the icosahedral I-$Al_{80}Mn_{20}$ and the decagonal T-$Al_{78}Mn_{22}$ quasicrystalline alloys. The zero-field resistivity $\rho(T,H=0)$, was measured in the range 0.4–50 K. Both samples exhibited a $-\log_{10}T$ dependence for $\rho(T,H=0)$ over a broad range of temperatures, followed by an approach to a constant value at the lowest temperature. The longitudinal magnetoresistance, $\rho_L(T,H)$, which was measured for magnetic fields H up to 75 kOe and for $T=1.5$ and 4.2 K, is positive for the I-phase sample and negative for the T-phase one. The transverse magnetoresistance, $\rho_T(T,H)$, is negative for both samples. The values measured for $\rho_L(T,H)$ and $\rho_T(T,H)$ are comparable to the ones measured in conventional metallic spin glasses.

I. INTRODUCTION

Of all the quasicrystalline alloys[1] discovered so far, the Al:Mn alloys show additional complexity because of their magnetic properties.[2–7] Despite the sharp x-ray diffraction peaks, a large number of experimental techniques have shown that the structural and physical properties of the Al:Mn quasicrystal samples resemble those of metallic and spin glasses.[2,4–9] Such a behavior is consistent with the lack of translational symmetry characteristic of quasicrystals which possess long-range orientational order only.[10]

In two previous communications we reported the heat capacity at constant magnetic field (C_H) of the icosahedral[4] I-$Al_{80}Mn_{20}$ and of the decagonal[6] T-$Al_{78}Mn_{22}$ alloys as a function of temperature T and applied magnetic field H down to low temperatures. The main results reported there were (1) no evidence was found for the anomalously high density of states at the Fermi energy predicted by a model for the electronic states of a quasicrystal, (2) an excess in the T^3 (phononlike) term was observed indicating the presence of low-energy modes as in metallic glasses, and (3) a broad peak whose maximum was centered around 1.0 K was observed. It was shown that the broad anomaly could not be associated with the spin-glass phase transition because the temperature of the observed maximum that occurs in C_H is the same for both samples, whereas their freezing temperatures T_g are much higher and are different for the two materials (in our sample T_g is 3.0 K for the I-$Al_{80}Mn_{20}$ and 7.8 K for the T-$Al_{78}Mn_{22}$). The lack of a dependence of H for the anomaly in T-$Al_{78}Mn_{22}$ suggests that its origin might be structural, such as a two-level system or tunneling states with an energy-dependent probability distribution of the kind observed in materials with a large number of defects.[11] This interpretation, however, is not unique. A Kondo system with a randomly distributed local field can yield similar results.[12] Both mechanisms give rise to a logarithmic temperature dependence for the resistivity, but they have a different behavior for the magnetoresistance.

In order to check these concepts and to investigate further the magnetism in these quasicrystalline alloys, we measured the resistivity (ρ) in zero applied magnetic field over the range $0.35 < T < 60$ K and the low T ($T=1.5$ and 4.2 K) longitudinal and transverse magnetoresistance (ρ_L and ρ_T) for H up to 75 kOe. We also performed measurements on the aluminum-rich alloy I-$Al_{83}Mn_{17}$ for comparison with previous reported results[13,14] and on icosahedral I-Al_6Ru for an estimates of the low-tempeature nonmagnetic residual resistivity. To the best of our knowledge there have been no prior results reported for quasicrystals having a similar sample composition, nor reports of a logarithmic temperature dependence for $\rho(T,H=0)$ over such a broad range in temperatures followed by saturation at lower temperatures.

There is at present active debate in the literature over the question of whether the icosahedral and decagonal materials one works with are really quasicrystals.[15] Questions of this type are beyond the scope of this paper. When we refer to our materials as quasicrystals, it is in the sense of following the current common usage; it is not intended to prejudge the question of whether the materials are quasicrystals in the formal sense.

II. SAMPLES AND MEASUREMENT PROCEDURES

The samples used in the present measurements are pieces of ribbon taken from the same batch of samples reported in Refs. 5 and 6. They were prepared by melt spinning in an argon atmosphere. X-ray diffraction and NMR measurements indicated that the I-$Al_{80}Mn_{20}$ and T-$Al_{78}Mn_{22}$ are more than 95% and 99% quasicrystalline

phase, respectively. Details on the preparation of the samples can be found elsewhere.[16]

All three resistivities were measured using the standard dc four-probe technique. The electrical leads to the sample were made of gold wire attached to the sample with silver paste. Great care was exercised in cleaning up the surface of the sample before making the contacts. The resistivity at $H=0$ for T down to 0.35 K was measured in a ^{3}He cryostat. Both ρ_L and ρ_T were measured in a ^{4}He cryostat equipped with a 80-kOe superconducting magnet. In the magnetoresistance measurement the change in the voltage across the sample as a function of H was continuously monitored as H was swept slowly. This condition is necessary in order to reduce the heating of the sample by eddy currents. Each sweep from $H=0$ up to 75 kOe took more than 30 min. Although the resolution of our experiment was of the order of 50 ppm for the change in resistance, the absolute resistivity was limited to $\pm 10\%$ because of the uncertainty in the dimensions of the samples.

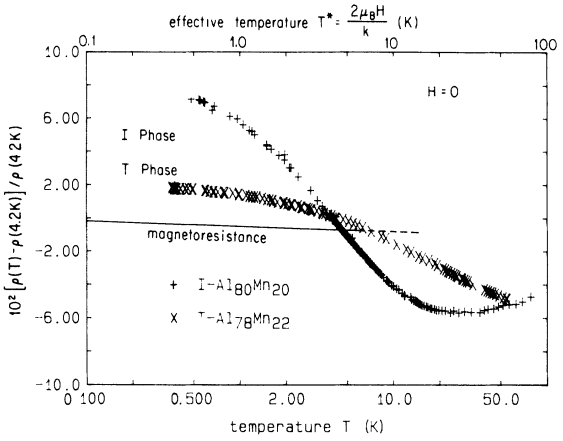

FIG. 1. Low temperature variation of the resistivity at $H=0$ for the I-$Al_{80}Mn_{20}$ and T-$Al_{78}Mn_{22}$ samples. The variation is plotted with respect to the resistivity at $T=4.2$ K, $\rho(4.2\text{ K})$. Values are listed in Table I. The solid line and scale at the top show the transverse resistance of the T-$Al_{78}Mn_{22}$ sample using an equivalent magnetic temperature scale.

III. EXPERIMENTAL RESULTS

The behavior of ρ for $H=0$ as a function of T for two samples is shown in Fig. 1 and summarized for all of them in Table I. There are several features in common. First, a rather large room-temperature resistivity is observed. Second, the residual resistance ratio, $\rho(300\text{ K})/\rho(4.2\text{ K})$, is of the order of the unity (for the I-$Al_{80}Mn_{20}$ and T-$Al_{78}Mn_{22}$ it is $\cong 0.8$). Finally, the temperature dependence of the I-$Al_{80}Mn_{20}$ and the T-$Al_{78}Mn_{22}$ shows a negative logarithmic temperature dependence, $-\log_{10}T$, for T below 10 and 50 K, respectively, followed by saturation in the low-temperature regime (see Fig. 1). Both samples show a crossover from the logarithmic to the saturation regime around $T_K \cong 4$ K. The room-temperature resistivity in the simple Drude model yields an electron mean free path l of 0.3, 1.1, and 1.5 Å for the T-$Al_{78}Mn_{22}$, I-$Al_{80}Mn_{20}$, and T-$Al_{83}Mn_{17}$, respectively. For the nonmagnetic I-Al_6Ru the electron mean free path is 2.8 Å.

We noticed small jumps in the absolute value of the resistivity and magnetoresistance at low temperatures. However, after normalizing $\rho(T,H=0)$ with respect to its value at 4.2 K, all the data points lie on the same curve. Because the jumps occur at random in temperature, we believe that they are more likely to be caused by poor electrical contact between grain boundaries in the quasicrystals, or breakdown and formation of magnetic clusters, rather than being associated with unusual features in the electronic density of states.[17] We also did not find a broad maximum or the power-law temperature dependence that is observed in the low-temperature resistivity of canonical metallic spin glasses.[18,19]

The behavior of the longitudinal magnetoresistance $\rho_L(T,H)$ for I-$Al_{80}Mn_{20}$, T-$Al_{78}Mn_{22}$, and I-$Al_{83}Mn_{17}$ is shown in Fig. 2 for $T=1.5$ and 4.2 K. The main result from these measurements is that the observed change in the resistivity $\Delta\rho_L$ with the applied magnetic field for these samples has the same order of magnitude as that reported for other magnetic alloys:[20] $\Delta\rho_L(T,H)/\rho_L(T,H=0)\cong 10^{-3}$. The measurements are also not strongly temperature dependent. Furthermore, $\Delta\rho_L(T,H)=\rho_L(T,H)-\rho_L(T,H=0)$ is positive for the I-phase samples and negative for the T-phase one. The transverse magnetoresistance, $\rho_T(T,H)$ is, however, negative for all three magnetic samples (Fig. 3) and positive for the nonmagnetic one, I-Al_6Ru. It is weakly dependent on T for the T-phase sample, but shows a somewhat stronger variation for the I-$Al_{80}Mn_{20}$ sample.

TABLE I. Measured resistivity and calculated conduction electron mean free path for several quasicrystal samples.

Alloy	ρ(300 K) ($\mu\Omega$ cm)	ρ(77 K) ($\mu\Omega$ cm)	ρ(4.2 K) ($\mu\Omega$ cm)	l(300 K) (Å)
I-$Al_{83}Mn_{17}$	128	118	119	1.5
I-$Al_{80}Mn_{20}$	177	210	228	1.1
T-$Al_{78}Mn_{22}$	613	672	725	0.3
I-Al_6Ru	69	42	35	2.8

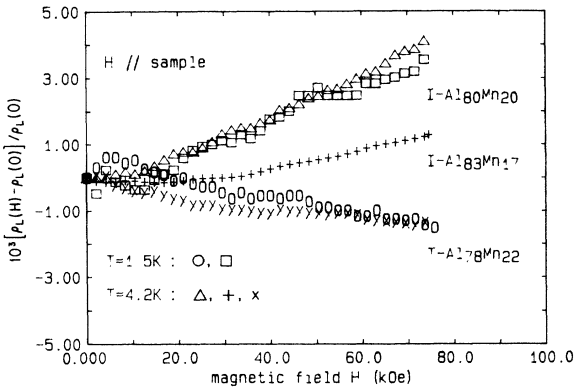

FIG. 2. Fractional variation of the longitudinal magnetoresistance, $[\rho_L(T,H)-\rho_L(T,H=0)]/\rho_L(T,H=0)$, for two quasicrystalline materials as a function of H at $T=1.4$ and 4.2 K. The zero-field values are listed in Table I.

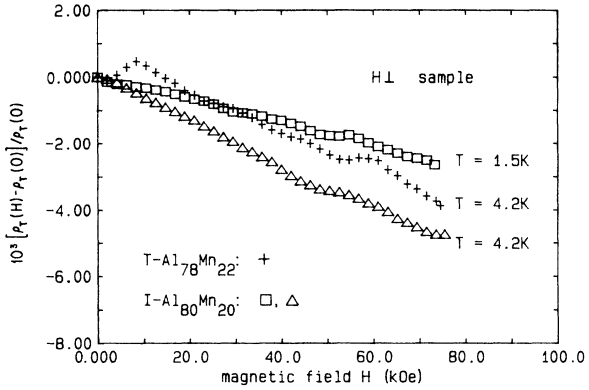

FIG. 3. Fractional variation of the transverse magnetoresistance, $[\rho_T(T,H)-\rho_T(T,H=0)]/\rho_T(T,H=0)$, for two quasicrystalline materials as a function of H at $T=1.4$ and 4.2 K. The zero-field values are listed in Table I.

IV. INTERPRETATION AND DISCUSSION

There are two "conventional" mechanisms which lead to a logarithmic temperature dependence of $\rho(T)$ with saturation at lower temperatures: (a) scattering by tunneling states or two-level systems associated with structural defects,[11] and (b) the Kondo effect, which is usually analyzed on the basis of conduction electron scattering by an isolated magnetic impurity. Because our materials show substantial magnetic interactions[4,6] among the magnetic impurities, we discuss our results in terms of a model for the magnetoresistance of metallic spin glasses.

A. Tunneling states or two-level systems

Three experimental observations suggest the two-level-system (TLS) model as a possible explanation for the resistivity temperature dependence: (1) The $-\log_{10}T$ and saturation behavior (Fig. 1), (2) the weak magnetic-field dependence on the resistivity which for $H=75$ kOe corresponds to a change of less than 1% of the total residual resistivity (Fig. 2), and (3) the lack of a field dependence observed on the heat capacity anomaly[6] of the T-$Al_{78}Mn_{22}$. In order to compare the number of TLS required to explain the large residual resistivity at low temperatures with that obtained from a fitting of the heat capacity to a Schottky anomaly, we estimate the residual resistivity based upon a model of resonant scattering of electrons by TLS in the unitary limit:[11]

$$\Delta\rho(T=0)\cong\pi\frac{5}{12}\frac{1}{\rho_0}\frac{\hbar m}{e^2}\frac{N_{\rm TLS}}{N}\ , \tag{1}$$

where m, e, and N are, respectively, the mass, charge, and total number of the conduction electrons, $\rho_0=mk_F/2\pi^2$ (k_F is the Fermi wave vector) is the conduction electron density at the Fermi energy, and $N_{\rm TLS}$ is the total number of active TLS's up to the characteristic crossover temperature T_K ($\cong 4$ K). Using our measured $\Delta\rho\cong 5\times10^{-7}$ Ω m for the T-$Al_{78}Mn_{22}$ sample and 1.7 Å for k_F one finds $N_{\rm TLS}/N\cong 4\times10^{-2}$. This number, which is a lower limit because of using the unitary limit, is at least one order of magnitude larger than that deduced from the specific-heat data. Hence, it is unlikely that conventional TLS are responsible for the anomalously high residual resistivity and its large logarithmic temperature dependence.

B. Kondo effect and the magnetoresistance of metallic spin glasses

The single impurity Kondo effect is not expected to work in this case because of magnetic interactions present in the spin-glass phase.[20] The importance of these interactions are evident in the H dependence[4,6] of C_H and from the magnetoresistance (Figs. 2 and 3) measurements. Furthermore, if the single-ion Kondo effect were responsible for $\rho(T)$ and $\rho_T(T,H)$, we would expect to see a variation in $\rho_T(T,H)$, that follows roughly $\rho(T,H=0)$ using an effective temperature $T^*=(T+2\mu_B H/k_B)$. The observed variation for T-$Al_{78}Mn_{22}$ is shown on Fig. 1 by the solid line using the scale at the top of the figure. It is clear that the 1% variation of $\rho_T(T^*)$ is much too small to match the corresponding 13% variation in $\rho(T)$. We therefore eliminate the single-ion Kondo effect as an explanation for the observed behavior.

There are, however, some features in the magnetoresistance of our samples which are common to metallic spin glasses. We consider some of them for comparison.

Because of the scatter in the data, the term in $\rho_T(T,H)$ that is quadratic in H cannot be obtained from our measurements. We can however, estimate the ratio of the quadratic coefficient $b(T,c)$ to the square of the s-d exchange potential, $J_{s\text{-}d}^2$, using the simplified expressions of Mookerjee for the spin-glass magnetoresistance,[20] the magnetic data given in Table II, and the results for the temperature dependence of the zero-field resistivity. We find $b(T,\bar{c})/J_{s\text{-}d}^2$ to be approximately 2.3×10^{-6}

TABLE II. Parameters for two quasicrystalline metallic spin glasses.

Parameter	I-$Al_{80}Mn_{20}$ (icosahedral)	T-$Al_{78}Mn_{22}$ (decagonal)
S	5/2	5/2
$\bar{S}$	5[a]	12[b]
c (at. %)	20	22
$\bar{c}$ (%)	0.2[a]	0.092[b]
p_{eff} (μ_B)[c]	11.0[a]	25.6[b]
T_g (K)	3.0[a]	7.8[b]
Θ (K)[c]	−1.4[a]	−1.5[b]
T_K (K)	4.1	4.0
n (10^{-28} m^{-3})	18.1[d]	
E_F (eV)	11.7[d]	
$b(T,\bar{c})/J_{s\text{-}d}^2$ (10^{-5} kOe^2 eV^{-2})	−0.23	1.5
$J_{s\text{-}d}$ (eV)	1.6	7.1
V (eV)	11	17

[a]Data from Ref. 4.

[b]Data from Ref. 6.

[c]p_{eff} and Θ are the effective moment and Neél temperature, respectively. Other symbols are defined in the text.

[d]Data from Ref. 21.

kOe^{-2}eV^{-2} and 1.5×10^{-5} kOe^{-2}eV^{-2} for the I-$Al_{80}Mn_{20}$ and for the T-$Al_{78}Mn_{22}$ samples, respectively. In this analysis, each cluster is treated as a single magnetic impurity with the large effective moment obtained from magnetization measurements.[4,6] The concentration of clusters ($\bar{c}$) and its effective spin ($\bar{S}$) is given in Table II. This table also includes for comparison entries that describe the metallic spin-glass properties. An estimate of $J_{s\text{-}d}$ can be made from its relation to the freezing temperature: $T_g \propto \bar{c}^{\,2/3} J_{s\text{-}d}$. On this basis we find $J_{s\text{-}d} \approx 1.6$ eV for I-$Al_{78}Mn_{22}$ and $J_{s\text{-}d} \approx 7.1$ eV for T-$Al_{78}Mn_{22}$. This large exchange potential is expected in part because of its dependence on the magnetic impurity spin S [$J_{s\text{-}d} \propto S(S+1)$]. Since $S=\frac{5}{2}$ for a single Mn^{3+} impurity, one finds that $\bar{S}$ is equivalent to a cluster of two and five Mn^{3+} ions for the I-phase and T-phase sample, respectively. Also the concentration of clusters in I-$Al_{80}Mn_{20}$ is only twice as large as in T-$Al_{78}Mn_{22}$. Consequently, the exchange potential per Mn atom is larger in T-$Al_{78}Mn_{22}$ than in I-$Al_{80}Mn_{20}$.

The zero-field high-temperature resistivity can also be used to obtain a value for the Coulomb potential V provided one knows the exchange potential[20] $J_{s\text{-}d}$:

$$\rho(H=0)=c\frac{3\pi m}{4\hbar e^2}\frac{1}{nE_F}[V^2+J_{s\text{-}d}^2 S(S+1)] , \qquad (2)$$

where m and e are the mass and charge of the electron, n is the density of electrons, E_F is the Fermi energy, and c and S are the concentration and spin of the magnetic atoms. By using the measured room temperature resistivity, the magnetic data given in Table II, and assuming that the values of n and E_F are the same as for the host metal, one obtains $|V| \approx 11$ eV and $|V| \approx 17$ eV for I-$Al_{80}Mn_{20}$ and for T-$Al_{78}Mn_{22}$, respectively.

In comparison with several metallic spin glasses[20] the values for the Coulomb potential of the quasicrystal samples are substantially larger. The values for the quasicrystals are, furthermore, rather different themselves. These differences suggest that the approach used to treat $\rho_T(T,H=0)$ as in conventional metallic spin glasses does not apply to our quasicrystal samples.

V. FURTHER DISCUSSION AND CONCLUSIONS

It is interesting to note that the large residual resistivity gives a very short electron mean free path (Fig. 1 and Table I), which according to the Ioffe-Regel criterion, appears to include the Al:Mn quasicrystalline alloys in the strong-localization limit. However, no theory we know of predicts in this limit a logarithmic temperature dependence for a three-dimensional system.[22] Attempts to explain the large residual resistivity with a model of Penrose tilings did not provide a quantitative agreement with the experiments.[23]

We have reported low-temperature measurements of the resistivity and high-field magnetoresistance in two previously investigated quasicrystalline alloys: the I-$Al_{80}Mn_{20}$ and the T-$Al_{78}Mn_{22}$. In our attempts to explain the results, we are unable to identify a unified theory that explains existing specific-heat and magnetization data and can account for the resistivity of I-$Al_{80}Mn_{20}$ and T-$Al_{78}Mn_{22}$. The interplay among structural disorder, spin-glass behavior, and possible intrinsic mechanisms associated with the long-range orientational order in these alloys creates a regime that requires further investiga-

tions in order to determine the mechanisms of their thermodynamic and electrical transport properties.

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