Mössbauer effect and electronic transport studies of icosahedral $Al_{50}Pd_{10}Mn_{25-x}Fe_xB_{15}$ alloys

M. Yewondwossen, J. E. Jagger, Z. Koziol, and R. A. Dunlap
Department of Physics, Dalhousie University, Halifax, Nova Scotia B3H 3J5, Canada

The temperature and applied magnetic field dependence of the resistivity of icosahedral $Al_{50}Pd_{10}Mn_{25-x}Fe_xB_{15}$ ($x=0$ and 5) alloys has been measured between 4.2 and 300 K. At low temperature, $T<30$ K, the resistivity showed a rapid decrease with increasing temperature and may be described by a combination of weak localization (WL) and magnetic scattering effects. At higher temperatures the resistivity is adequately described by the temperature dependence of the structural and magnetic effects as described by Boltzmann-type transport. The measured magnetoresistance of both samples is consistent with theoretical predictions based on WL. The room temperature Mössbauer effect spectrum of the $x=5$ sample showed a well resolved doublet with mean isomer shift (relative to room temperature α-Fe) and quadrupole splitting of $+0.22$ mm/s and 0.36 mm/s, respectively. These results indicate that the Fe probe nuclei do not carry a magnetic moment in these alloys. © *1996 American Institute of Physics.* [S0021-8979(96)16508-0]

I. INTRODUCTION

Quasicrystalline (QC) alloys which contain Mn have shown a wide variety of magnetic behavior and, in some cases have exhibited varying degrees of magnetic order. Typically, these magnetically ordered materials have been characterized by small Mn magnetic moments and low Curie temperatures. Recently, alloys which exhibit high Curie temperatures and substantial Mn moments have been reported[1–5] in the Al-Mn-Pd-B system. The availability of these materials allows for the investigation of the relationship of magnetic order and quasicrystallinity. In the present work we report on magnetotransport and Mössbauer effect studies of $Al_{50}Pd_{10}Mn_{25-x}Fe_xB_{15}$ alloys.

II. EXPERIMENTAL METHODS

Samples of $Al_{50}Pd_{10}Mn_{25-x}Fe_xB_{15}$ ($x=0$ and 5) were prepared by melt spinning. Resulting ribbons were shown to be single phase icosahedral quasicrystals by x-ray diffraction studies. The transverse magnetoresistance was measured at temperatures from 4.2 to 300 K in applied fields up to 5.5 T. The room temperature ^{57}Fe Mössbauer effect spectrum of the $x=5$ sample was obtained using a conventional constant acceleration drive system.

III. RESULTS AND DISCUSSION

The magnetoresistance data of $Al_{50}Pd_{10}Mn_{25-x}B_{15}Fe_x$ QC alloys ($x=0$, 5) are shown in Fig. 1. The most general expression for the magnetoresistance, $\Delta\rho(B)$, due to Fukuyama and Hoshino[6] which includes the effects of spin-orbit scattering, Zeeman splitting, and magnetic impurities is given by;

$$\frac{\Delta\rho}{\rho}=\rho A\sqrt{\frac{eB}{\hbar}}\left\{\frac{1}{2\sqrt{1-\gamma}}\left[f_3\left(\frac{B}{B_-}\right)-f_3\left(\frac{B}{B_+}\right)\right]-f_3\left(\frac{B}{B_2}\right)-\sqrt{\frac{4B_{so}}{3B}}\left(\frac{(\sqrt{t_+}-\sqrt{t_-})}{\sqrt{1-\gamma}}+\sqrt{t}-\sqrt{t+1}\right)\right\} \quad (1)$$

where

$$A=\frac{e^2}{2\pi^2\hbar},$$

$$\gamma=\left(\frac{3g^*\mu_B B}{8eD(B_{so}-B_s)}\right)^2,$$

$$B_\phi=B_i+2B_s,$$

$$B_2=B_i+\tfrac{2}{3}B_s+\tfrac{4}{3}B_{so},$$

$$t=\frac{3B_\phi}{4(B_{so}-B_s)},$$

$$B_\pm=B_\phi+\tfrac{2}{3}(B_{so}-B_s)(1\pm\sqrt{1-\gamma})+2B_s,$$

$$t_\pm=t+\tfrac{1}{2}(1\pm\sqrt{1-\gamma}),$$

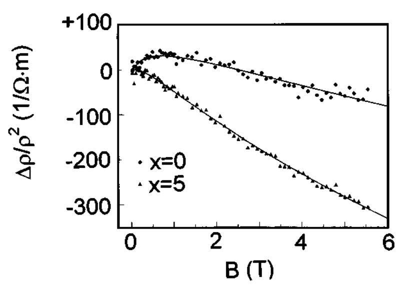

FIG. 1. The magnetoresistivity at 4.2 K of $Al_{50}Pd_{10}Mn_{25-x}B_{15}Fe_x$ plotted as a function of B. The solid lines are fits to WL theory.

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TABLE I. Fitted parameters values for $Al_{70-x}Pd_{15}Mn_{15}B_x$.

Alloy (x)	$1/\tau_i$ ($\times 10^9$ s^{-1})	$1/\tau_{so}$ ($\times 10^9$ s^{-1})	$1/\tau_s$ ($\times 10^9$ s^{-1})	ρ(fitted) ($\mu\Omega$ cm)	ρ(exp) ($\mu\Omega$ cm)
0	5.4	10	0.7	870	1700
5	7.8	0.45	0.1	1200	1400

and D is the diffusion coefficient. The characteristic fields are related to electron scattering times through relations of the type

$$B_x = \frac{\hbar}{4eD\tau_x}, \quad (2)$$

where $x=i$, so, and s refer to the inelastic, spin-orbit, and magnetic spin-flip scattering times, respectively. The Kawabata function, $f_3(x)$,[7] has been written in the form proposed by Baxter *et al.*;[8]

$$f_3(x) = 2\left(\sqrt{2+\frac{1}{x}} - \sqrt{\frac{1}{x}}\right) - \left[\left(\frac{1}{2}+\frac{1}{x}\right)^{-1/2} + \left(\frac{3}{2}+\frac{1}{x}\right)^{-1/2}\right] + \frac{1}{48}\left(2.03+\frac{1}{x}\right)^{-3/2}. \quad (3)$$

The magnetoresistance due to weak localization, $\Delta\rho_{WL}$, as given by Eq. (1), is negative in the case of weak spin-orbit scattering systems, i.e., $\tau_i < \tau_{so}$. In the case of strong spin-orbit scattering systems the magnetoresistance is positive and $\tau_i > \tau_{so}$.

The fitting procedure adopted for the samples studied is as follows. The WL contribution to the magnetoresistance is fitted with the temperature-dependent inelastic scattering time $\tau_i(T)$, and the temperature independent scattering times τ_{so} and τ_s as free parameters. The diffusion coefficient is taken to be 0.075 cm^2 s^{-1},[9] from literature values for similar quasicrystals. Quantum corrections to the resistivity predict $\Delta\rho/\rho \propto \rho$. Using ρ as a free parameter in the WL expression allows for a determination of the resistivity in a way that is independent of the sample geometry.[10]

The agreement between WL theory and the experimental data is good over the entire range of fields as illustrated by the fits shown in Fig. 1. The magnetoresistance of $x=0$ sample is positive in region $B<2.5$ T followed by a negative magnetoresistance in the remaining range of field. This feature reflects the moderate spin-orbit scattering case in which $\tau_i \approx \tau_{so}$. For the $x=5$, the magnetoresistance is totally negative and reflects the presence of weak spin-orbit scattering, $\tau_i < \tau_{so}$. The value of the fitted parameters are given in Table I.

The temperature dependence of the resistivity is illustrated in Fig. 2. The resistivity of the $Al_{70-x}Pd_{15}Mn_{15}B_x$ QC alloys is 1700 and 1400 $\mu\Omega$ cm at 4.2 K (for $x=0$ and 5, respectively). Various theories have been proposed to interpret the high values of the resistivity of QC's (e.g., Refs. 11–13). In general, two effects dominate $\rho(T)$ in QCs at low temperature; weak localization and magnetic scattering. The resistivity behavior at low temperature in $Al_{70-x}Pd_{15}Mn_{15}B_x$ is primarily due to magnetic scattering effects. This assertion may be illustrated as follows: The low temperature resistivity can be written as;

$$\rho(T) \approx \Delta\rho_{WL}(\tau_l(T), \tau_{so}, \tau_s, D) + \rho_{Kondo}, \quad (4)$$

where $\Delta\rho_{WL}$ and ρ_{Kondo} are the WL and Kondo contributions, respectively. Using the scattering times described above the WL component may be subtracted from the resistivity data. The remaining data exhibit a $\ln(T)$ dependence for all the samples as is expected for Kondo-type scattering. At higher temperatures the resistivity is adequately described by the temperature dependence of Boltzmann-type transport.

The room temperature ^{57}Fe Mössbauer effect spectrum of icosahedral $Al_{50}Pd_{10}Mn_{20}Fe_5B_{15}$ is illustrated in Fig. 3. This spectrum shows a well defined doublet. The analysis of such spectra has been considered in detail by a number of authors (e.g., Refs. 14–16). In cases where the splitting is quadrupole in nature, spectra may be analyzed on the basis of a quadrupole distribution or a set of discrete splittings. Evidence seems to suggest that in related QC's two distinct classes of transition metal sites exist.[14,17,18] An analysis of

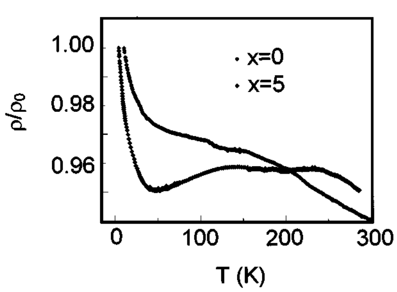

FIG. 2. Temperature dependence of the electrical resistivity normalized to the value at 4.2 K for $Al_{50}Pd_{10}Mn_{25-x}Fe_xB_{15}$ ($x=0$, 5).

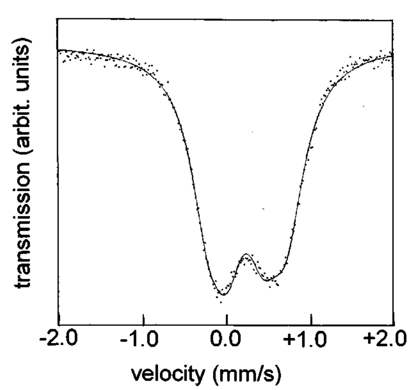

FIG. 3. Room temperature ^{57}Fe Mössbauer effect spectrum of icosahedral $Al_{50}Pd_{10}Mn_{20}Fe_5B_{15}$. The solid line is a fit to two symmetric doublets.

TABLE II. Room temperature ^{57}Fe Mössbauer effect parameters for the spectrum of icosahedral $Al_{50}Pd_{10}Mn_{20}Fe_5B_{15}$. Quadrupole splittings, Δ, and isomer shifts, δ, (relative to α-Fe) are given for the two doublets as described in the text. Mean parameter values are also given.

	Δ (mm/s)	δ (mm/s)
doublet No. 1	0.233	+0.219
doublet No. 2	0.493	+0.230
mean value	0.362	+0.224

Mössbauer spectra in terms of two discrete sites is illustrated in Fig. 3 and are summarized in Table II. In this case the two doublets have been chosen to have similar isomer shifts. Mean quadrupole parameters are consistent with Mössbauer effect results obtained for similar QCs. Spectra have also been fitted on the basis of a combined Zeeman and quadrupole interaction. The quality of fit for this analysis did not show any improvement over the previous analysis, suggesting that the Fe probe atoms do not carry magnetic moments in these alloys. There has been considerable work on Fe moment formation in QCs and evidence seems to suggest that Fe atoms prefer to reside in sites where a moment does not form.[14,19] The present measurements are consistent with this picture.

IV. CONCLUSIONS

The temperature and applied magnetic field dependence of the resistivity of icosahedral $Al_{50}Pd_{10}Mn_{25-x}Fe_xB_{15}$ ($x=0$ and 5) alloys has been measured. At low temperature results may be explained by a combination of weak localization and Kondo-type magnetic scattering effects. At higher temperatures the resistivity is adequately described by the temperature dependence of Boltzmann-type transport. The room temperature Mössbauer effect spectrum of the $x=5$ sample showed a well resolved doublet. A goodness-of-fit analysis indicated that the spectrum was appropriately described by a combination of two symmetric quadrupole doublets rather than an analysis based on the presence of a Zeeman splitting. This analysis suggests that the Fe probe atoms prefer to reside in sites where they do not carry a magnetic moment.

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