Electrical resistivity and point contact studies on Al–Pd–Mn icosahedral quasicrystals

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Abstract. We report measurement of the electrical resistivity $\rho(T)$ and point contact differential conductance (dI/dV) for icosahedral $Al_{70}Pd_{30-x}Mn_x$ ($x = 7, 9$ and 11) at low temperatures. Below 10 K, for $x = 11$, $\rho(T)$ follows a log T dependence. All the three compositions show a sharp minimum in the differential conductance near zero bias. Magnitude of the dip in the point contact conductance increases as the Mn concentration increases. The dip near zero bias also follows a log V dependence for $V < 30$ mV.

Keywords. Electrical resistivities; point contact studies; Al–Pd–Mn.

The icosahedral quasicrystals (i-QC), having self-similar and aperiodic arrangement of atoms, give rise to interesting electronic and magnetic properties (Kimura and Takeuchi 1991; Poon 1992). A number of thermodynamically stable Al-based transition metals i-QC have been discovered. The magnetic property of 3d transition metals and alloys are very sensitive to the local atomic configuration in the system. The localized magnetic moments are mainly affected by this local arrangement of the atoms. The study of the influence of the icosahedral symmetry on the transition metal atoms has been a topic of research in the recent years (Shinohara *et al* 1992). Al–Pd–Mn alloy belongs to the i-QC, having magnetic moments. The presence of magnetic moments gives rise to interesting behaviour of resistivity $\rho(T)$ at low temperatures. Recently the magnetic properties of polycrystalline Al–Pd–Mn i-QC have been investigated (Matsuo and Nakano 1992; Chernikov *et al* 1993; Hattori *et al* 1994). It has been found that the icosahedral phase possess local magnetic moments in the Mn atomic sites and the value of the moment strongly depends on Mn concentration (Hattori *et al* 1994).

In this article we report the temperature-dependence of electrical resistivity $\rho(T)$ and point-contact studies at liquid helium temperature on Al–Pd–Mn i-QC alloys. In $\rho(T)$ measurement, energy of the electrons interacting with the impurity can only vary by an amount $\sim k_B T$. Whereas, in point-contact studies at liquid helium temperature one obtains information about the interaction of conduction electrons in the range of eV, where eV can be greater than $k_B T$ or eV $\ll k_B T$. This feature greatly increases the range of applicability of point-contact spectroscopy (PCS) for studying the interaction of conduction electrons not only with phonon, but also with a wide range of other elementary excitation, and for studying scattering of electrons by impurities and defects. Point-contact spectra measurement refers to the measurement of the current–voltage characteristics and of dI/dV and d^2I/dV^2 as a function of the bias voltage (V) of a micro-contact junction made between the tip and a sample. Among the various features which are obtained in the dI/dV curve, of particular interest is the zero-bias

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anomaly observed in the alloys. The structure in dI/dV can be related to scattering of electrons by magnetic impurities (D' Ambrumenil and White 1982; D'Ambrumenil *et al* 1983).

The $Al_{70}Pd_{30-x}Mn_x$ ($x = 7, 9$ and 11) alloys were prepared from high purity (99·999%) constituent elements by induction melting in pure argon atmosphere. The QCs were made from the ingot by melt spinning on a copper wheel in a ribbon form of thickness $\sim 25 \mu$m. The resistivity of the ribbons were measured by low frequency (20 Hz) ac four-probe method (Banerjee and Raychaudhuri 1994) in the temperature range 4·2 K–300 K. The point-contact measurements were done at 4·2 K with a elec-trochemically etched gold tip. The details of the experiment are given elsewhere (Srikanth and Raychaudhuri 1991, 1992). X-ray powder diffraction pattern obtained from as-quenched samples for the three compositions of the alloy could be indexed by all odd or all even numbers, indicating icosahedral quasilattice is F-type (FCI) icosahedral phase without any other crystalline or quasicrystalline phase.

The temperature dependence of the resistivity $\rho(T)$ of the three samples ($x = 7, 9, 11$) are shown in figure 1. All the compositions show negative temperature coefficient of resistivity (TCR) at high temperature ($T > 100$ K), like other conventional quasi-crystals. All of them show a maximum in ρ for $T < 100$ K. At low temperatures these alloys show interesting features in $\rho(T)$ as a function of Mn concentration. We observe that for $x = 11$, ρ increases steeply with decrease in temperature below 20 K with an inflection point at about 40 K. This sudden increase in ρ is not observed for $x = 7$ and 9 for temperature down to 4·2 K. The maximum in ρ shifts towards higher temperature as the concentration of Mn increases. The absolute value of $\rho(300$ K) decreases with the increase in Mn concentration. Figure 2 shows the results of point-contact studies on the three alloys ($x = 7, 9$ and 11). We have plotted the normalized conductance

$$G(V)/G(V = 0) \ (= (d\bar{I}/dV)/(dI/dV)_{V=0}),$$

for 20 Ω junction at 4·2 K. The curves have been shifted for the sake of clarity. All the three compositions show a sharp decrease in the conductance near zero bias. It can be
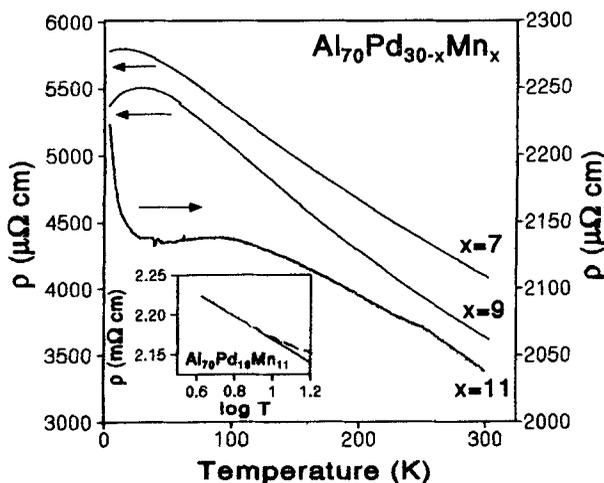

Figure 1. The electrical resistivity as a function of temperature for the three alloys $Al_{70}Pd_{30-x}Mn_x$ ($x = 7, 9$ and 11). In the inset we show for $x = 11$ in a log T scale.

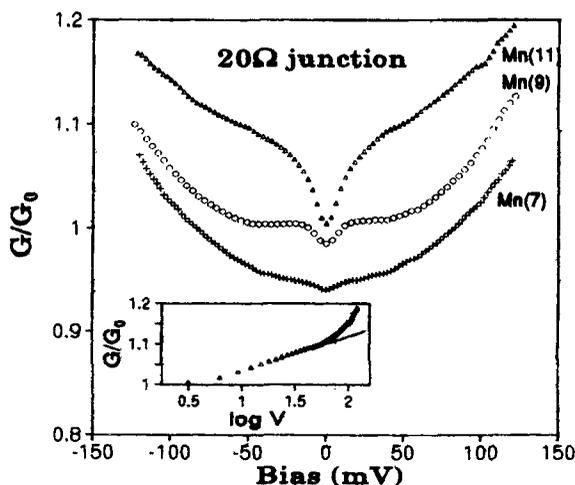**Figure 2.** The normalized conductance $G(V)/G(V=0)$ $(=(dI/dV)/(dI/dV)_{V=0})$ for $20\,\Omega$ junction at $4\cdot2\,\text{K}$ for $Al_{70}^{-}Pd_{30-x}Mn_x$ ($x=7,9$ and 11). In the inset we show for $x=11$ in a log V scale.

seen that the magnitude of the dip in the conductance increases as the Mn concentration increases.

The temperature-dependence of resistivity is found to be very sensitive to Mn concentration. The negative TCR at high temperature can be attributed to either (a) weak-localization effects (Alt'shuler and Aronov 1985) or (b) diffraction effect from the temperature-dependence of the structure factor (Cote and Meisel 1981). A detailed discussion can be found in Banerjee *et al* (1995). A resistivity maxima is observed in this alloy, which can be attributed to weak-antilocalization due to spin-orbit interaction effects (Matsuo *et al* 1994; Banerjee *et al*, to be published). This feature has been observed in many stable Al-based transition metal *i*-QC alloys like Al–Cu–Fe (Haberkern *et al* 1993) and Al–Cu–Cr (Banerjee *et al* 1995; Banerjee *et al*, to be published). The shift of the maxima towards higher temperature as the Mn concentration increases can be attributed to the onset of spin-orbit interaction or spin scattering at higher temperature for greater Mn concentration.

Recently it has been shown that *i*-QC Al–Pd–Mn alloys show a spin glass behaviour at low temperatures (Chernikov *et al* 1993; Hattori *et al* 1994). The spin freezing temperature T_f increases linearly as the Mn concentration increases. For $x=11$ the steep increase in ρ below $20\,\text{K}$ can be attributed to spin glass transition. The inset of figure 1 shows ρ as a function of log T. It can be clearly seen that ρ follows a log T behaviour for $T<10\,\text{K}$ as observed in Kondo systems (dilute magnetic alloys). We do not observe this steep rise in ρ for $x=7$ and 9 till $4\cdot2\,\text{K}$ because the T_f is $<4\cdot2\,\text{K}$. The above discussion qualitatively explains the temperature-dependence of ρ as a function of Mn concentration.

The zero-bias feature i.e. minimum in dI/dV (or maximum in dV/dI) has been observed in various noble metals (Cu, Au) doped with magnetic impurities (Mn) (D'Ambrumenil *et al* 1983). The system Al–Pd–Mn can be approximated to magnetically dilute systems, since the system has very low atomic percentage of magnetic Mn atoms (1–3% of all Mn atoms) (Hattori *et al* 1994). Moreover, the system shows a spin-glass transition at low temperatures (Hattori *et al* 1994). The important difference to note between this system and

the systems studied previously (Cu:Mn, Cu:Fe, etc) is that the resistivity of these i-QC alloys are orders of magnitude more than the conventional dilute magnetic systems. The notable feature here is that the zero-bias anomaly shows a systematic variation with the Mn concentration; the depth increases with increase in Mn concentration. This is in contrast to that observed in (Au, Cu):Mn where the minimum in conductance at zero bias split into two minimum with the increase in Mn concentration. For dilute magnetic alloys, the scattering time has been evaluated theoretically (Suhl 1973; Hamann 1976), the theory predicts a similar behaviour for the temperature-dependence of the bulk resistivity and voltage-dependence of the inverse of the scattering time (τ^{-1}(eV)). Experimentally, one observes this behaviour for noble metals doped with magnetic impurities. Taking clue from this, we have plotted for the case of 11% Mn the $G(V)$ vs log V in the inset of figure 2. We can see that the dip near the zero bias follows a log V dependence up to $V = 30$ mV. More qualitative understanding is necessary in relating this voltage as an equivalent temperature. We also observe in figure 2 that the conductance increases with increase in bias voltage in contrast to a decrease in the case of Cu:Mn alloys (D'Ambrumenil and White 1982; D'Ambrumenil 1983). This may be due to increase in electrical conductivity with increase of temperature (negative TCR for $T > 100$ K) unlike in (Au, Cu):Mn alloys. It can be seen from figure 2 that the voltage at which the conductance shows a sudden decrease, decreases with decrease in Mn concentration. This may be attributed to the temperature at which the steep rise in the resistivity occurs (the spin freezing temperature), also decreases with decrease in Mn concentration.

Thus, qualitatively we can correlate the resistivity and the point-contact conductance data in this system. To determine the size of the internal fields etc the point-contact study has to be carried out under magnetic field, and also a systematic study of the conductance curves as a function of temperature is essential.

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