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The distinction between the magnetic properties of quasicrystalline and amorphous $Al_{85-x}Pd_{15}Mn_x$ alloys

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Abstract. Quasicrystalline and amorphous $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys have been prepared by rapid melt quenching and high-rate DC sputtering. The saturation magnetic moment M_s , the magnetic Mn atom ratio, the magnetic moment per magnetic Mn atom P_c and spin glass behaviour have been investigated for both these phases.

The temperature dependence of DC magnetic susceptibility for these quasicrystalline and amorphous alloys follows a Curie-Weiss Iaw over a wide temperature range. The value of M_s and the average magnetic moment per Mn atom $\langle P_c \rangle$ for the quasicrystalline alloys are much smaller than those for the amorphous alloys. The magnetic Mn atom ratio in the quasicrystalline phase is about 10%, being almost half that in the amorphous phase in a similar manner to the $Al_{80-x}Cu_{20}Mn_x$ (x = 13, 14 and 15) alloy system. The Mn concentration dependence of P_c in the quasicrystalline phase is steeper than that in the amorphous phase, and the value of P_c of the quasicrystalline $Al_{70}Pd_{15}Mn_{15}$ alloy exceeds $5\mu_B$ of a bare moment of Mn. It is, therefore, considered that a giant moment is formed by polarization of 4d electrons of Pd in the quasicrystalline alloy.

The present alloys in both phases are magnetically dilute alloys and spin glass behaviour has been observed. The spin freezing temperature $T_{\rm f}$ for the quasicrystalline phase is lower than those for the amorphous phase in accordance with the smaller Mn atom ratio. From these results, a marked difference in the magnetic properties between the quasicrystalline and amorphous phases has been confirmed, reflecting their different local atomic structure.

1. Introduction

A number of icosahedral quasicrystalline Al-based alloys have been developed over a period of several years (Shechtman *et al* 1984, Sainfort and Dubost 1986, Tsai *et al* 1987, 1990) and various physical properties have been investigated (Suck 1993, Akiyama *et al* 1993). On the other hand, the structure of amorphous alloys has often been connected with icosahedral clusters (Steinhardt *et al* 1983, Sachdev and Nelson 1984), and some similarity of short-range atomic arrangement between quasicrystalline and amorphous phases has been expected. In fact, some structural resemblance has been confirmed by x-ray structure analysis of quasicrystalline and amorphous $Al_{75}Cu_{15}V_{10}$ alloys (Matsubara *et al* 1988). However, the physical properties of quasicrystalline alloys would be different from those of their amorphous counterparts, reflecting their different local atomic structure.

It is well known that the magnetic properties of 3d transition metals and alloys are very sensitive to their atomic structures, and the localized magnetic moments are mainly affected by the kinds of nearest-neighbour atom, the atomic distance and the coordination number. In particular, the magnitude of localized magnetic moment in various Mn alloys and intermetallic compounds is closely correlated to the Pauling valence (Mori and Mitsui 1968). Therefore, a distinction of magnetic properties between quasicrystalline and amorphous alloys is expected. Accordingly, the magnetic properties of quasicrystalline and amorphous Al-Mn alloys have been investigated (Fukamichi *et al* 1987, Goto *et al* 1988). However, no clear distinction was confirmed in this system because the localized moment is hardly formed in the Al matrix (Grüner 1974). On the other hand, a clear difference in the magnetic properties between quasicrystalline and amorphous phases has been confirmed in Al_{80-x}Cu₂₀Mn_x (x = 12, 13, 14 and 15) alloys. In this system, the magnetic properties of quasicrystalline alloys are weaker than those of their amorphous counterparts (Fukamichi *et al* 1993). It is well known that the formation of a localized magnetic moment is much easier in a Cu matrix than in an Al matrix for magnetic dilute alloy systems (Heeger 1969, Grüner 1974). Therefore, the substitution of Cu for Al makes clear the difference in magnetic properties between the two phases, reflecting the difference in the local atomic structure.

Recently, it has been reported that icosahedral quasicrystalline Al-Pd-Mn alloys are formed by melt quenching in wide concentration ranges (Tsai et al 1990, 1991). A structural identification of the quasicrystalline alloys has been carried out by x-ray and neutron diffraction (Boudard et al 1991, 1992, 1993, de Boissieu et al 1994). The local atomic structures such as the atomic distance and the coordination number, which closely correlate with the magnetic properties, for an icosahedral quasicrystalline Al₇₁Pd₁₉Mn₁₀ alloy have been investigated by EXAFS above the K absorption edges of Al, Pd and Mn (Sadoc and Dubois 1993). In the present paper, therefore, the magnetic properties of icosahedral quasicrystalline and amorphous $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys have been investigated. Taking into consideration the local atomic structure mentioned above, the difference in the magnetic properties between these phases will be discussed. In dilute crystalline alloys such as Au-Fe and Cu-Mn, spin glass behaviour caused by the RKKY interaction has often been observed below about 10% Mn or Fe (Cannella and Mydosh 1972, Coles et al 1975). In addition, the 4d conduction electrons of Pd are polarized by magnetic impurities, resulting in a giant magnetic moment in dilute crystalline Pd-TM (TM = Fe, Co and Mn) alloys (Bozorth et al 1961, Chouteau and Tournier 1971, Star et al 1975). The concentration of magnetic Mn atoms in the present alloys is very low, and hence these alloys should be considered as magnetically dilute alloys. Therefore, spin glass behaviour and giant magnetic moment in the present $Al_{85-r}Pd_{15}Mn_r$ (x = 12, 13, 14 and 15) alloys are also discussed.

2. Experimental details

Alloying was performed by arc melting in an Ar gas atmosphere purified with a Ti getter, using starting materials of 99.99 wt.% pure Al, 99.99 wt.% Pd and 99.9 wt.% Mn. Quasicrystals were prepared by rapid melt quenching using a single-roller apparatus. The composition of targets for sputtering was adjusted so as to obtain the same composition as that of the melt quenched samples. Bulk amorphous alloy samples about 0.3 mm thick were prepared by high-rate DC sputtering for 3 d on a water cooled Cu substrate. The Ar gas pressure during sputtering was 40 mTorr and the target voltage was 1.0 kV. The samples were removed from the Cu substrate by mechanical polishing or with a solution of H_2SO_4 (25 cm³) + H_2O (1000 cm³) + Cr_2O_3 (500 g) at about 350 K. The quasicrystalline and amorphous states were confirmed by x-ray diffraction using Cu K α radiation. The compositional analysis was carried out by an inductively coupled plasma (ICP) method, and

the analysed compositions in the two phases were in agreement with each other within about 0.1–0.2 at.%. The magnetization up to 55 kOe, the temperature dependence of DC magnetic susceptibility and the magnetic cooling effect were measured with a SQUID magnetometer (Quantum Design). The very high-field magnetization measurements up to 340–400 kOe were made with a pulse magnet. The temperature dependence of AC magnetic susceptibility was measured by a mutual induction method in 10 Oe with a frequency of 80 Hz.

3. Results and discussion

Figures 1(a) and (b) respectively shows the temperature dependence of magnetic susceptibility for quasicrystalline and amorphous $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys. Over a wide temperature range, except very low temperatures, the magnetic susceptibility follows a Curie–Weiss law in a similar manner to $Al_{100-x}Mn_x$ and $Al_{80-x}Cu_{20}Mn_x$ alloys (Goto *et al* 1988, Fukamichi *et al* 1987, 1991, 1993), and to a single-grained quasicrystalline $Al_{68}Pd_{23}Mn_9$ alloy (Matsuo *et al* 1993). The Curie constant *C* is obtained from the following equation:

$$\chi - \chi_0 = C/(T - \theta_{\rm P}) \tag{1}$$

where χ_0 is the temperature independent susceptibility and θ_p the paramagnetic Curie temperature. All samples exhibit a negative value of θ_p ranging from -2 to -15 K, suggesting that the antiferromagnetic Mn-Mn exchange interactions are dominant in both these phases.



Figure 1. (a) The temperature dependence of DC magnetic susceptibility for quasicrystalline (Q) $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys. (b) The temperature dependence of DC magnetic susceptibility for amorphous (A) $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys.

In the localized magnetic moment systems, the magnetic carrier ratio of the Rhodes-Wohlfarth plot is unity (Rhodes and Wohlfarth 1963) and the Curie constant is connected with the average magnetic moment per Mn atom $\langle P_c \rangle$ as given by the following expression:

$$C = N\mu_{\rm B}^2 \langle P_{\rm c} \rangle (\langle P_{\rm c} \rangle + 2)/3k_{\rm B}$$
⁽²⁾

where N is the number of Mn atoms per mole, μ_B the Bohr magneton and k_B the Bohrzmann constant. The concentration dependence of $\langle P_c \rangle$ for the quasicrystalline and amorphous



Figure 2. The concentration dependence of the average magnetic moment per Mn atom $\langle P_c \rangle$ for quasicrystalline (Q) and amorphous (A) Al_{85-x}Pd₁₅Mn_x (x = 12, 13, 14 and 15) alloys.

Al_{85-x}Pd₁₅Mn_x (x = 12, 13, 14 and 15) alloys is shown in figure 2. For both these phases $\langle P_c \rangle$ increases with increasing Mn content, but the magnitude of $\langle P_c \rangle$ in the quasicrystalline phase is lower than that in the amorphous phase. Furthermore, the concentration dependence of the former is steeper than that of the latter. Similar trends have also been observed in the Al_{80-x}Cu₂₀Mn_x (x = 12, 13, 14 and 15) system (Fukamichi *et al* 1993). It has been reported that the saturation magnetic moment M_s is much smaller than $\langle P_c \rangle$ for the quasicrystalline Al_xMn_{100-x} ($14 \leq x \leq 22$) and Al_{80-x}Cu₂₀Mn_x (x = 12, 13, 14 and 15) alloys (Goto *et al* 1988, Fukamichi *et al* 1991, 1993), indicating the coexistence of magnetic and non-magnetic Mn atoms. It has also been confirmed that there are magnetic and non-magnetic Mn atoms in quasicrystalline Al-Mn (Fe)-Si alloys by the Mössbauer effect (Eibschutz *et al* 1987, Edagawa *et al* 1987).

The magnetic Mn atom ratio, x_m/x , is estimated from the following expression based on the assumption that the g factor is two:

$$M_{\rm s}((x/x_{\rm m})M_{\rm s} + 2\mu_{\rm B}) = 3k_{\rm B}C/N.$$
(3)

In order to obtain the ratio, x_m/x , the saturation magnetic moment M_s is required. Figure 3 displays the high-field magnetization curves up to 400 kOe at 4.2 K for the quasicrystalline and amorphous phases. The magnetization curves exhibit a marked curvature, and the saturation is not enough to determine M_s even in such a high magnetic field H. Then, as shown in figure 4, the value of M_s is deduced by fitting 1/H against magnetization in a similar manner to a 1/H scaling plot of the magnetization for dilute crystalline Cu-Mn alloys (Bellissent *et al* 1987). The value of M_s for the quasicrystalline Al₇₃Pd₁₅Mn₁₂ alloy



Figure 3. High-field magnetization curves up to 400 kOe at 4.2 K for quasicrystalline (Q) and amorphous (A) $AI_{85-x}Pd_{15}Mn_x$ (x =12, 13, 14 and 15) alloys. Quasicrystalline, (A) x = 12, (B) x = 13, (C) x = 14, (D) x = 15; amorphous. (a) x = 12, (b) x = 13, (c) x = 14, (d) x = 15.

is estimated to be 4.3 emu g⁻¹, as given by the arrow. The concentration dependence of M_s thus obtained is shown in figure 5, together with the results for icosahedral and decagonal quasicrystalline Al₇₀Pd₁₄Mn₁₆ alloys (Satoh et al 1994) whose compositions are slightly different from those of the present alloy system. Comparing with the data in figure 1, the concentration dependence of M_s in both quasicrystalline and amorphous phases is related in the same way as $\langle P_c \rangle$. The important point to note is that the magnitude of M_s is much smaller than that of $\langle P_c \rangle$ for both phases. Similar results have been obtained for other Albased alloy systems (Fukamichi et al 1991, 1993). In spite of the different concentrations of Pd and Al, the value for the icosahedral Al₇₀Pd₁₄Mn₁₆ alloy (*) is very close to the extended line of the quasicrystalline $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys, and the value for the decagonal phase (x) is much smaller than that of the icosahedral phase. Using M_s obtained above, the value of x_m/x is calculated from (3). Figure 6 shows the concentration dependence of the magnetic Mn atom ratio, x_m/x , together with that of the $AI_{80-x}Cu_{20}Mn_x$ alloy system (Fukamichi et al 1993), for comparison. In both $Al_{85-x}Cu_{20}Mn_x$ (x = 12, 13, 14 and 15) and $Al_{80-x}Cu_{20}Mn_x$ (x = 12, 13, 14 and 15) systems, x_m/x of the quasicrystalline alloys is about 10%, being about half that of the amorphous alloys. The value of x_m/x for quasicrystalline Al–Mn and Al–Mn–Si alloys has been estimated to be 3 \sim 10% from low-temperature specific heat data (Lasjaunias et al 1987, Berger et al 1988a, b), which is comparable to the present results. An estimation of x_m/x for an icosahedral Al₇₀Pd₂₁Mn₉ alloy by magnetization measurements has been reported to be about 1% (Chernikov et al 1993), which is one order of magnitude smaller than the present results. However, it should be pointed out that a linear extrapolation of the reciprocal magnetic field for magnetization around 50 kOe is inadequate, in contrast to



Figure 4. The high-field magnetization against the reciprocal magnetic field of quasicrystalline (Q) $Al_{73}Pd_{15}Mn_{12}$ alloy, together with the scaling plot of dilute crystalline (C) Cu-Mn alloys. The arrow indicates the corresponding magnetization.



Figure 5. The concentration dependence of the saturation moment M_s for quasicrystalline (Q) and amorphous (A) Al_{85-x}Pd₁₅Mn_x (x = 12, 13, 14 and 15) alloys, together with the data for icosahedral (i) and decagonal (D) quasicrystalline Al₇₀Pd₁₄Mn₁₆ alloys.

figure 4, resulting in an underestimation of M_s that leads to such a small value of x_m/x .

The local atomic structures, i.e., the atomic distance and the coordination numbers, of an icosahedral quasicrystalline Al₇₁Pd₁₉Mn₁₀ alloy have been investigated using the extended x-ray absorption fine structure (EXAFS) above the K absorption edges of Al, Pd and Mn (Sadoc and Dubois 1993) and confirmed no existence of Mn–Mn pair nearest neighbours.



Figure 6. The concentration dependence of the ratio of magnetic Mn atoms, x_m/x , for quasicrystalline (Q) and amorphous (A) $AI_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys, together with that of $AI_{80-x}Cu_{20}Mn_x$ (x = 12, 13, 14 and 15) alloys.

As mentioned before, the average magnetic moment $\langle P_c \rangle$, the saturation magnetic moment M_s and the magnetic Mn atom ratio x_m/x in the quasicrystalline phase are smaller than those in the amorphous phase. These results could be explained by such a local atomic structure. That is, in the quasicrystalline $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys, $\langle P_c \rangle$, M_s and x_m/x are small because the Mn-Mn pairs are considered to be rare or there are no such pairs, as in the quasicrystalline $Al_{71}Pd_{19}Mn_{10}$ alloy. These values should be larger in the amorphous alloys because the probability of developing the Mn-Mn pairs would increase owing to the random distribution of atoms. EXAFS experiments for the amorphous $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys are highly desired to discuss this in more detail.

The average localized moment of magnetic Mn atoms, P_c , is estimated from the following expression:

$$P_{\rm C} = (x/x_{\rm m})M_{\rm s}/\mu_{\rm B}.\tag{4}$$

The concentration dependence of P_c is similar to those of $\langle P_c \rangle$ and M_s . The magnitude of P_c for the quasicrystalline Al₇₀Pd₁₅Mn₁₅ alloy, however, is larger than that for the amorphous counterpart, as shown in figure 7, and more noteworthy is that the value exceeds $5\mu_B$ of a bare Mn moment. Larger magnetic moments have also been obsreved in the icosahedral quasicrystalline Al₇₀Pd_{30-x}Mn_x (x = 16, 17 and 18) alloys (Satoh *et al* 1994). It is, therefore, concluded that these quasicrystalline alloys have a so-called giant magnetic moment in analogy with dilute crystalline Pd-TM (TM: Fe, Co and Mn) alloys (Bozorth *et*



Figure 7. The concentration dependence of the magnetic moment per magnetic Mn atom, P_c , for quasicrystalline (Q) and amorphous (A) $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys.

al 1961, Chouteau and Tournier 1971, Star et al 1975). As is well known, in these dilute crystalline alloys, the spin polarization of 4d electrons of Pd induced by the magnetic element brings about a giant magnetic moment. The increase of P_c with increasing concentration has also been reported for dilute crystalline alloy systems (Crangle and Scott 1965). In contrast to dilute crystalline Pd–Fe and Pd–Co alloys, quasicrystalline Al₇₀Pd₁₆Fe₁₄ and Al₇₀Pd₁₀Co₂₀ alloys carry no giant magnetic moment (Fukamichi et al 1991).

The atomic percentage of magnetic Mn atoms for both phases is 1-3% by total content, so they should be regarded as magnetically dilute alloys. Therefore, the presence of spin glass behaviour due to the fluctuations of the RKKY interaction are expected in the present $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys. The spin freezing temperature T_f of a quasicrystalline Al₇₁Pd₂₀Mn₉ alloy has been reported to be 0.5 K from AC magnetic susceptibility measurement (Chernikov et al 1993). The spin glass state in this alloy is caused by the fluctuations of the RKKY interaction because it is considered that the nearest-neighbour Mn-Mn pair is scarcely formed in a similar manner to the quasicrystalline $Al_{71}Pd_{19}Mn_{10}$ alloy mentioned before. Figures 8(a) and (b) displays the zero-field cooled (ZFC) and field cooled (FC) magnetizations of quasicrystalline $Al_{85-x}Pd_{15}Mn_x$ (x = 13, 14 and 15) and amorphous $Al_{75-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys, respectively, in a field of 30 Oe. It exhibits a clear hysteresis between ZFC and FC magnetizations at low temperatures. The temperature dependence of AC magnetic susceptibility in 10 Oe at 80 Hz for these alloys also exhibits a cusp as shown in figure 9. From these results, it is concluded that these alloys are spin glasses. We should not overlook the fact that the temperature dependence of the magnetic susceptibility given in figure 1 becomes less

divergent at very low temperatures because of the existence of the spin glass state. Dilute crystalline Pd-Mn alloys also exhibit spin glass behaviour (Coles *et al* 1975. Ho *et al* 1982). It should be emphasized that the magnetic properties of the quasicrystalline $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys are similar to those of dilute crystalline Pd-Mn alloys because both systems have a giant magnetic moment and exhibit spin glass behaviour. Shown in figure 10 is the concentration dependence of the spin freezing temperature T_f obtained from the cusp temperature of the AC magnetic susceptibility. Both these phases exhibit a linear concentration dependence with the smaller ratio of the magnetic Mn atoms given in figure 5.



Figure 8. (a) The temperature dependence of the zero-field cooled (ZFC) and field cooled (FC) magnetizations for quasicrystalline (Q) $Al_{85-x}Pd_{15}Mn_x$ (x = 13, 14 and 15) alloys in a field of 30 Oe. (b) The temperature dependence of the zero-field cooled (ZFC) and field cooled (FC) magnetizations for amorphous (A) $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys in a field of 30 Oe.

A comparison of magnetic properties between icosahedral (*) and decagonal (×) quasicrystalline $Al_{70}Pd_{30-x}Mn_x$ (x = 16, 17 and 18) alloys has been made in the same concentration range (Satoh *et al* 1994). The value of M_s for the icosahedral quasicrystalline $Al_{70}Pd_{14}Mn_{16}$ alloy is larger than that for the decagonal counterpart as shown in figure 5. Other values of magnetic properties of the icosahedral quasicrystalline alloys are larger than those of the decogonal quasicrystalline alloys. In the present study, the magnitude of $\langle P_c \rangle$, M_s and T_f for the amorphous $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys is larger than that for the quasicrystalline alloys as seen from figures 2, 5 and 10. Therefore, although the compositions of these alloy systems are slightly different, it is concluded that $\langle P_c \rangle$, M_s



Figure 9. The temperature dependence of AC magnetic susceptibility of quasicrystalline (Q) and amorphous (A) $Al_{85-x}Pd_{15}Mn_x$ alloys in a field of 10 Oe with a frequency of 80 Hz. The arrows indicate the spin freezing temperature T_f .



Figure 10. The concentration dependence of the spin freezing temperature $T_{\rm f}$ for quasicrystalline (Q) and amorphous (A) Al_{85-x}Pd₁₅Mn_x (x = 12, 13, 14 and 15) alloys.

and $T_{\rm f}$ decrease in the sequence amorphous > icosahedral quasicrystalline > decagonal quasicrystalline alloys.

4. Conclusion

Quasicrystalline and amorphous $Al_{85-x}Pd_{15}Mn_x$ (x = 12, 13, 14 and 15) alloys have been prepared in order to investigate the difference in magnetic properties, which reflects the different local atomic structure. The main results are summarized as follows.

(i) The magnitude of the average magnetic moment per Mn atom $\langle P_c \rangle$ and the saturation magnetic moment M_s for the quasicrystalline alloys are smaller than those for the amorphous alloys.

(ii) The magnetic Mn atom ratio, x_m/x , for the quasicrystalline alloys is about half that for the amorphous alloys. This trend is very similar to that of $Al_{80-x}Cu_{20}Mn_x$ (x = 13, 14 and 15) alloys.

(iii) The concentration dependences of $\langle P_c \rangle$, M_s , x_m/x and the magnetic moment per magnetic Mn atom P_c for the quasicrystalline phase are steeper than those for the amorphous phase.

(iv) The quasicrystalline $AI_{70}Pd_{15}Mn_{15}$ alloy has a giant magnetic moment induced by polarization of 4d conduction electrons of Pd.

(v) Both phases exhibit spin glass behaviour and the spin freezing temperature $T_{\rm f}$ in the quasicrystalline phase is lower than that in the amorphous phase, reflecting the lower content of the magnetic Mn atoms.

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