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Magnetic Moment and Spin Glass Behavior of AlCuMn and AlPdMn Quasicrystalline and Amorphous Alloys*

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Synopsis

In the present review, the effective magnetic moment, the magnetic Mn atom ratio, partial substitution of TM and the spin glass behavior of AlCuMn, AlPdMn and AlMn quasicrystalline and amorphous alloys are discussed. Distinct differences in the magnetic properties between quasicrystalline and amorphous states have been confirmed in AlCuMn and AlPdMn alloy systems in contrast with those in AlMn alloys. The effective magnetic moments of AlCuMn and AlPdMn quasicrystalline alloys are smaller than those of amorphous counterparts, but much larger than those of AlMn alloys in the same concentration range.

The ratios of magnetic Mn atoms in AlCuMn and AlPdMn quasicrystalline alloys are about one half that of the amorphous counterparts, although there is no essential distinction in AlMn alloy systems. The spin glass behavior has been confirmed in AlCuMn amorphous alloys and AlPdMn quasicrystalline and amorphous alloys even below 15 %Mn, although AlMn alloys exhibit the spin glass behavior above 20 %Mn. These differences mentioned above can be explained by considering the difference in the forming ability of the localized magnetic moment among three alloy systems and in the local structure between the quasicrystalline and amorphous states. It should be noted that $Al_{70}Pd_{15}Mn_{15}$ quasicrystalline alloy has a giant magnetic moment in analogy with PdMn crystalline dilute alloys.

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Introduction

Various Al-based quasicrystalline alloys have been developed during several years. As well-known, it is not so easy for almost all 3d transition metals to have the localized magnetic moment in Al in the very dilute concentration range[1]. Therefore, the study on the formation of the localized magnetic moment in Al-based quasicrystalline alloys is of interest.

Magnetic properties of AlMn quasicrystalline, amorphous and crystalline alloys have been investigated systematically[2,3]. It should be noted that the former two alloys exhibit essentially the same tendencies in the concentration dependences of the effective magnetic moment and the spin freezing temperature [3,4]. On the other hand, the crystalline counterparts show no spin glass behavior and the value of the effective magnetic moments are about half those of the former alloys[2]. It has been pointed out from the data on the Mössbauer effect[5], NMR spectra[6]and the high-field magnetization[7,8] that there are magnetic and non-magnetic Mn sites in AlMn and its ternary quasicrystalline alloys such as AlMnSi and AlMnFe. The ratio of the magnetic Mn atoms in AlMn quasicrystalline alloys is essentially the same as that of the amorphous counterparts. From these results it is concluded that the magnetic properties of AlMn quasicrystalline alloys are very similar to those amorphous counterparts. Therefore, we can find no distinction of these properties between AlMn quasicrystalline and amorphous alloys.

Recently, new systems of AlCuMn and AlPdMn quasicrystalline alloys have been developed by Tsai et al[9-11]. These two and AlMn quasicrystalline alloys have Mackay icosahedral(MI) type structures[12]. It has been pointed out that $Al_{65}Cu_{20}Mn_{15}$ quasicrystalline alloy has the largest magnetic susceptibility, compared with that of other $Al_{65}Cu_{20}TM_{15}$(TM: transition metal) quasicrystalline alloys such as $Al_{65}Cu_{20}Fe_{15}$ and $Al_{65}Cu_{20}Cr_{15}$[13]. The susceptibility of AlPdTM quasicrystalline alloys shows a similar tendency. In general, the magnetic properties are very sensitive to the local environment such as the atomic distance and the coordination number. Especially, it has been demonstrated that the magnitude of magnetic moment of various manganese compounds is governed by the Pauling valence correlated with the local environment[14,15].

The detailed comparison of the magnetic and electrical properties of AlMn alloys between the quasicrystalline and amorphous states was already reported in the preceding paper[4]. Therefore, in the present review, recent results of AlCuMn and AlPdMn alloys in the quasicrystalline and amorphous states are presented and compared with those of AlMn alloys in these two states. The following five subjects are discussed:

(A) Effective Magnetic Moment
(B) Saturation Magnetization
(C) Ratio of Magnetic Mn Atoms
(D) Partial Substitution of TM
(E) Spin Glass Behavior

A) Effective Magnetic Moment

The magnetic properties of Fe group impurity in Cu and in Al have been investigated intensively from the standpoint of virtual bound state[1]. The Fermi energy of Al is 12 eV and the band width Γ in the virtual bound state is about 4eV. On the other hand, these values are respectively about 7 eV and 2 eV in Cu. Strictly speaking, the condition of the formation of localized magnetic moment is expressed by the Anderson hamiltonian but conventionally given by[1]

$$\Gamma < p\Delta E \qquad (1),$$

where ΔE is the energy deference between parallel and anti-parallel spin pairs, being about 0.6 ~ 0.8 eV, and p is the electron number in d-shell. Therefore, it is clear that the formation of localized magnetic moment in Al is very difficult but easy in Cu. Especially, Mn in Cu, + and - spin states are situated below and above the Fermi surface, respectively. Therefore, the localized magnetic moment is easily established, showing a Curie-Weiss type temperature dependence.

Figures 1 and 2 show the temperature dependence of susceptibility of AlCuTM and AlPdTM(TM:Transition Metal) quasicrystalline alloys. Both quasicrystalline alloys containing Mn show a remarkable temperature dependence, compared with other alloys. Figure 3 shows the temperature dependence of susceptibility of $Al_{65}Cu_{20}Mn_{15}$ and $Al_{70}Pd_{15}Mn_{15}$ quasicrystalline and amorphous alloys. From this figure, the susceptibility of quasicrystalline alloys is smaller than that of amorphous counterparts. The effective magnetic moment is deduced from the Curie constant C given by the following conventional equation;

$$\chi - \chi_o = C/(T-\theta_p) \qquad (2),$$

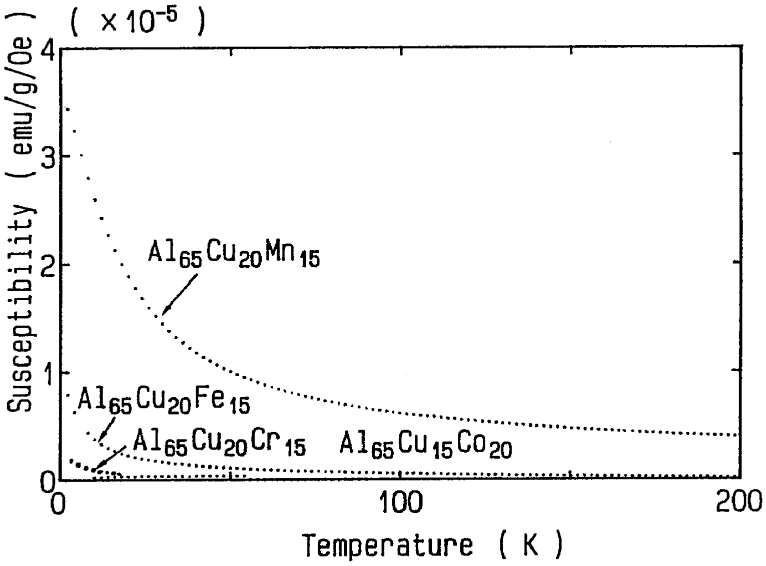

Fig.1 Temperature dependence of susceptibility of AlCuTM (TM: Mn, Fe, Co and Cr) quasicrystalline alloys.

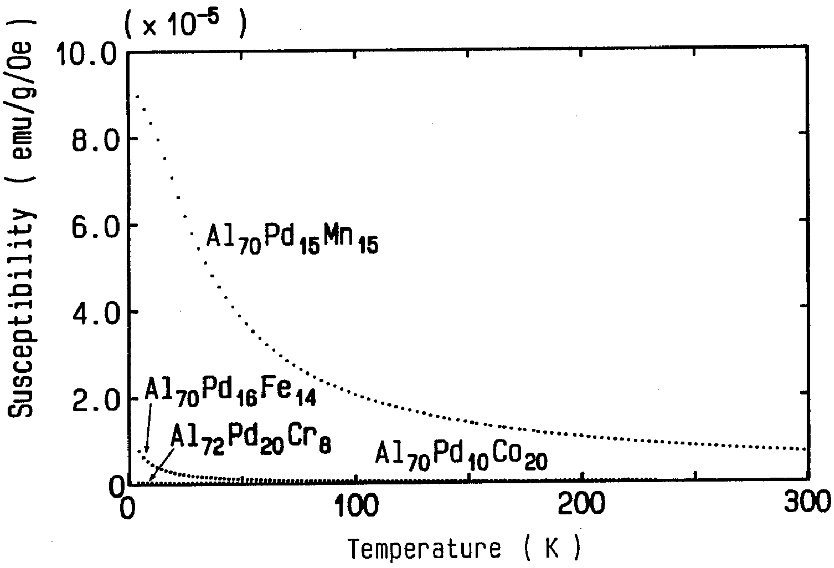

Fig.2 Temperature dependence of susceptibility of AlPdTM quasicrystalline alloys.

where χ_o and θ_p are the temperature independent susceptibility and the paramagnetic Curie temperature, respectively.

Figure 4 shows the concentration dependence of the effective magnetic moment of AlCuMn and AlPdMn alloys in the quasicrystalline and amorphous states, together with that of AlMn alloys in two states. AlMn alloys show the same tendency and the almost same value in both

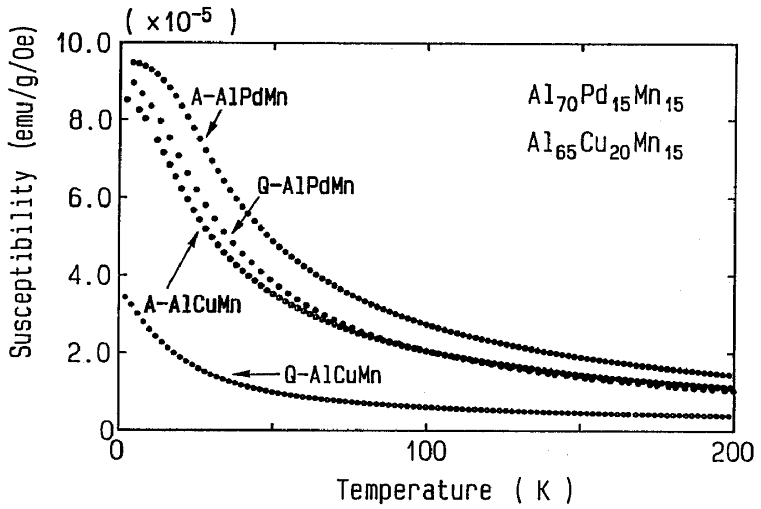

Fig.3 Temperature dependence of susceptibility of $Al_{65}Cu_{20}Mn_{15}$ and $Al_{70}Pd_{15}Mn_{15}$ alloys in the quasicrystalline(Q) and amorphous(A) states.

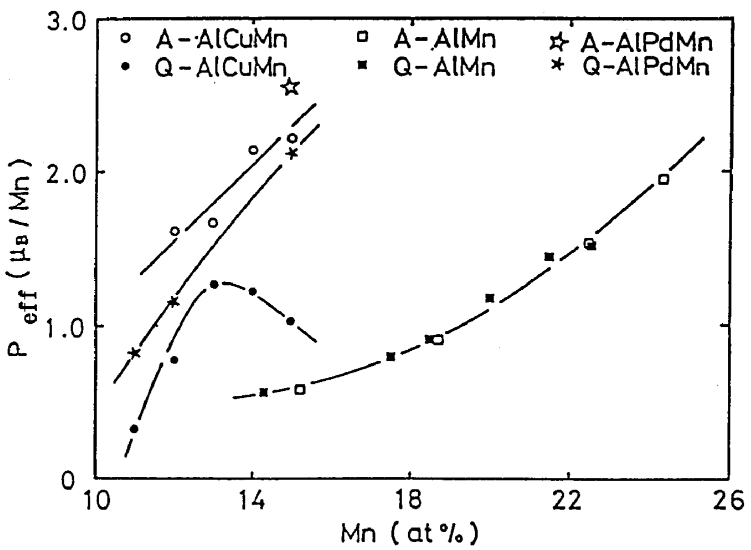

Fig.4 Concentration dependence of the effective magnetic moment of $Al_{100-x}Mn_x$, $Al_{80-x}Cu_{20}Mn_x$ and $Al_{85-x}Pd_{15}Mn_x$ alloys in the quasicrystalline(Q) and amorphous(A) states.

states[4]. The value of AlCuMn quasicrystalline alloys exhibits a broad maximum around 13 %Mn but the amorphous counterparts show a monotonic increase. Furthermore, the magnitude of the amorphous alloys is much larger than that of the quasicrystalline alloys[16]. The value of AlPdMn amorphous alloys is also larger than that of the quasicrystalline alloys[17].

B) Saturation Magnetization

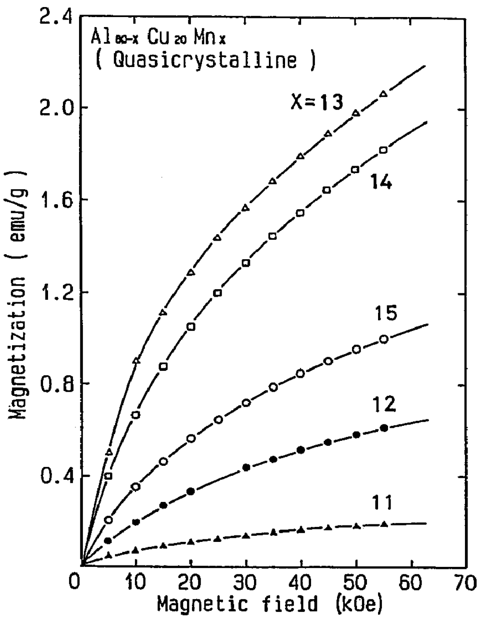

Fig.5(a) Magnetization curves at 4.2 K for $Al_{80-x}Cu_{20}Mn_x$ quasicrystalline alloys.

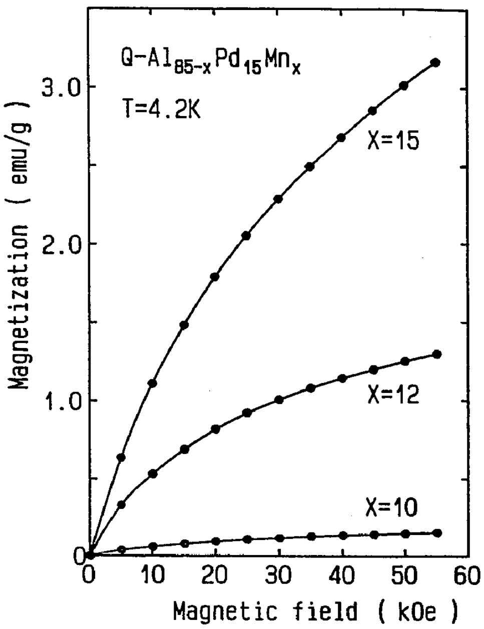

Fig.5(b) Magnetization curves at 4.2 K for $Al_{85-x}Pd_{15}Mn_x$ quasicrystalline alloys.

It has been pointed out that magnetization is much smaller than the value expected from the effective magnetic moment. This fact suggests that the magnitude of moment is not uniform. From various experiments, it has been confirmed that there are magnetic and non-magnetic Mn atoms[5-8]. Figures 5(a) and (b) show the magnetization curves up to 55 kOe measured by a SQUID magnetometer at 4.2 K for AlCuMn and AlPdMn quasicrystalline alloys, respectively. These alloys exhibit a strong curvature. In order to estimate the saturation magnetization M_s, the magnetization was measured up to 380 KOe in the pulsed magnetic fields. Figure 6 shows these data on $Al_{65}Cu_{20}Mn_{15}$ and $Al_{70}Pd_{15}Mn_{15}$ quasicrystalline alloys[16,17]. In such high fields the saturation is not enough to determine the saturation magnetization. By assuming a 1/H law, the value M_s of AlMnSi quasicrystalline alloy has been estimated by scaling with the data on CuMn crystalline dilute alloys[7]. In the present study, the values of M_s are deduced by using the same method. The representative 1/H plot for AlPdMn quasicrystalline alloy is given in Fig.7, together with that of CuMn dilute crystalline alloys[7]. The scaling seems to work well over wide range of the magnetic field. Concentration dependence of the saturation magnetization per Mn atom thus obtained for AlCuMn

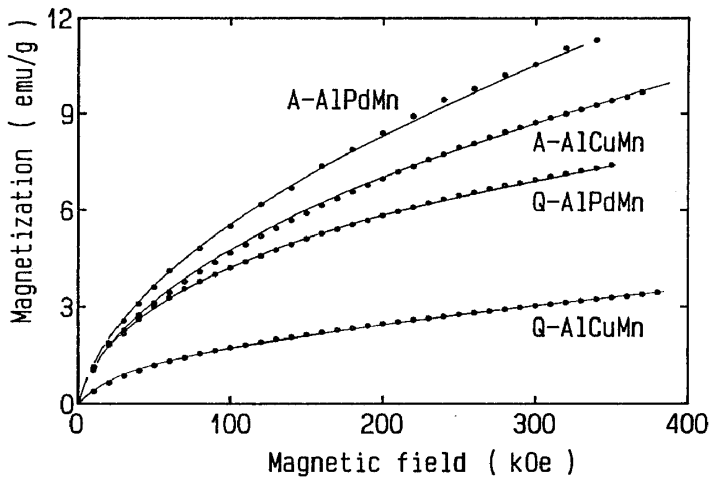

Fig.6 Magnetization curves of $Al_{65}Cu_{20}Mn_{15}$ and $Al_{70}Pd_{15}Mn_{15}$ quasicrystalline(Q) and amorphous(A) alloys measured up to 380 KOe at 4.2 K.

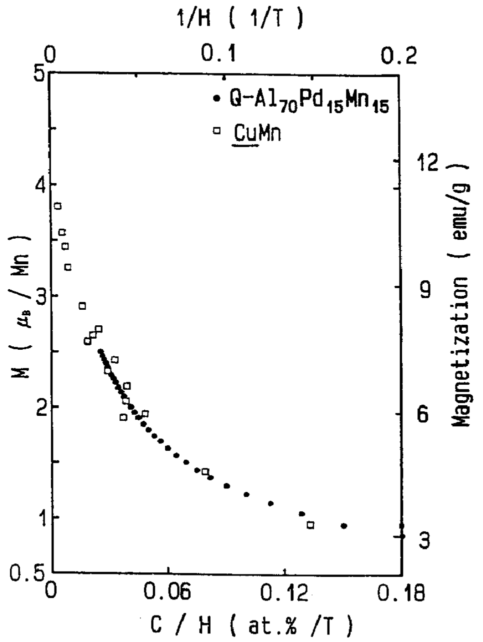

Fig.7 1/H plots of CuMn[7] and $Q\text{-}Al_{70}Pd_{15}Mn_{15}$.

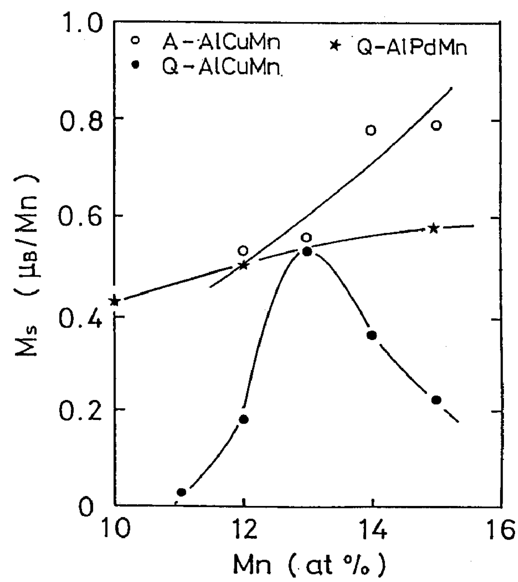

Fig.8 Concentration dependence of Q- and $A\text{-}Al_{80-x}Cu_{20}Mn_x$ and $Q\text{-}Al_{70}Pd_{30-x}Mn_x$.

quasicrystalline, amorphous alloys and AlPdMn quasicrystalline alloys is shown in Fig.8. The values of AlCuMn amorphous and AlPdMn quasicrystalline alloys vary linearly with increasing Mn content, but the value of AlCuMn quasicrystalline alloys exhibits a broad maximum around 13 %Mn in a similar manner as that of the effective magnetic moment as shown in Fig.4.

C) Ratio of Magnetic Mn Atoms

The saturation magnetization per Mn atom is much smaller than the effective magnetic moment as seen from Figs.4 and 8. These facts mean that there are magnetic and non-magnetic Mn atoms in those alloys. The ratio of magnetic Mn atoms, x_m/x, is correlated with the following expression;

$$M_s(M_s x/x_m + 2\mu_B) = 3k_B C/N \qquad (3),$$

where M_s and N are the saturation magnetization per Mn atom and the number of Mn atoms, respectively. The g value is assumed to be 2. The ratio of magnetic Mn atoms in three alloy systems is presented in Table 1. The values of AlCuMn and AlPdMn amorphous alloys are the

Table 1 Ratio of magnetic Mn atoms and mean magnetic moment of magnetic Mn atoms in Q- and A-states.

Alloy	Ratio of magnetic Mn atoms(%)	Mean magnetic moment of magnetic Mn atoms(μ_B/Mn)
Q-$Al_{85}Mn_{15}$	~ 24	~ 0.7
A-$Al_{85}Mn_{15}$	~ 24	~ 0.5
Q-$Al_{65}Cu_{20}Mn_{15}$	8.9	2.5
A-$Al_{65}Cu_{20}Mn_{15}$	18.7	4.2
Q-$Al_{70}Pd_{15}Mn_{15}$	9.3	6.3
A-$Al_{70}Pd_{15}Mn_{15}$	18.7	5.2

same and smaller than those of AlMn amorphous and quasicrystalline alloys. On the other hand, the values of the former two quasicrystalline alloys are almost same and about one half those of amorphous counterparts. The magnetic moment of magnetic Mn atoms is also calculated from Eq.(3). These values are also given in the same table. The value of $Al_{65}Cu_{20}Mn_{15}$ quasicrystalline alloy is about one half that of the amorphous counterpart. In the case of $Al_{70}Pd_{15}Mn_{15}$ quasicrystalline alloy, the value exceeds the largest value for the bare moment on the Mn atoms of 5 μ_B. Therefore, it is concluded that this alloy has a giant magnetic moment. As well-known, in the crystalline dilute alloys such as PdCo, PdFe and PdMn, the giant magnetic moments have been observed. Such as the giant magnetic moment in Pd dilute alloys is established due to the strong exchange interaction between the conduction electrons[18]. In the dilute alloys mentioned above, the spin polarization of the conduction electrons is induced by the magnetic elements. However, in the present study, AlPdCo and AlPdFe quasicrystalline alloys carry no large magnetic moment, having no giant magnetic moment. Therefore, only Mn atoms in the AlPdMn alloys have a giant magnetic moment.

In AlCuMn and AlPdMn quasicrystalline alloys, the ratios of the magnetic Mn atoms are much smaller but the magnetic moments are oppositely larger than those of the amorphous counterparts, respectively. These facts suggest that there is a distinct difference in the local structure between the quasicrystalline and amorphous states because the giant moment polarization is remarkably reduced with increasing atomic distance between the magnetic atom and the surrounding Pd atoms[18,19]. That is, it is expected that the magnetic atoms in the former state are surrounded by many Pd and Cu atoms, compared with those in the amorphous state. The structure of amorphous alloys has been connected with the icosahedral structure[20,21], and the structural similarity and the relationship between the $Al_{75}Cu_{15}V_{10}$ icosahedral quasicrystalline alloy and its amorphous counterpart have been discussed[22]. It has been reported that the pair distribution functions in the icosahedral and amorphous phases of $Pd_{58.8}U_{20.6}Si_{20.6}$ alloy are similar up to the second-nearest neighbors[23]. On the other hand, detailed structural analyses of AlMn alloys in the quasicrystalline and amorphous states were made through EXAFS measurements and the difference in the pair distribution function between these two states has been demonstrated[24]. In the crystalline state, almost all magnetic transition metals hardly carry the localized magnetic moment in Al in the dilute concentration range because the Fermi energy of Al is very high, compared with other host metals such as Cu and Au[25]. In such a circumstance, therefore, it is expected that no distinct difference is observed in AlMn quasicrystalline and amorphous alloys. That is to say, the magnetic properties are insensitive to the different local environment in AlMn alloys between the quasicrystalline and amorphous states. On the other hand, in Cu and Pd in the crystalline state, many magnetic transition metals easily carry the localized magnetic moment. Therefore, the slight difference in the local environment between the quasicrystalline and amorphous states would be sensitively reflected in the magnetic properties, although the present alloys contain only 20 %Cu or 15 %Pd.

D) Partial Substitution of TM

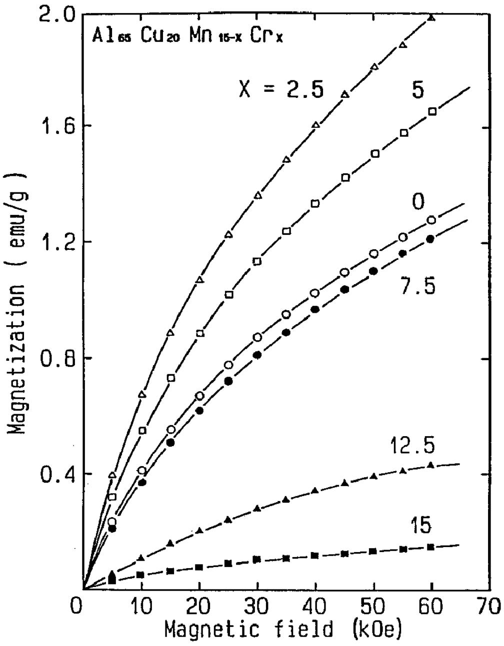

Fig.9 Magnetization curves of $Al_{65}Cu_{20}Mn_{15-x}Cr_x$ quasicrystalline alloys(4.2 K).

As mentioned above, the magnetic properties are very sensitive to the change of the local environment. Therefore, it is expected that these properties are affected by the partial substitution of the transition metal. Figure 9 shows the magnetization curves of Al-CuMnCr quasicrystalline alloys. In spite of the formation no localized moment in AlCuCr quasicrystalline alloys as seen from Fig.1, the magnetization is strongly increased at first by the partial substitution of Cr for Mn. Similar tendency has been confirmed in AlMnCr, AlCuMnV, AlCuMnMo and AlCuMnRu quasicrystalline alloys[28]. On the other hand, the magnetizations of AlCuMnNb and AlCuMnZr quasicrystalline alloys

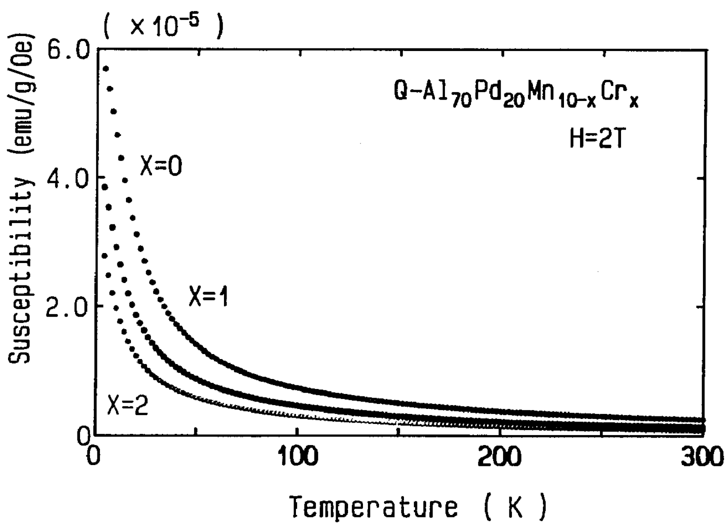

Fig.10 Temperature dependence of susceptibility of $Q\text{-}Al_{70}Pd_{20}Mn_{10-x}Cr_x$ alloys.

show a monotonic decrease with increasing substitutional element. In the case of AlPdMnCr alloys, there is no increase in the susceptibility as seen from Fig.10. This fact suggests that the magnetic moment of Mn atoms is saturated because they have a giant magnetic moment. Then, the magnetic moment is increased no longer, resulting in the decrease of the magnetization. Figure 11 shows the relationship between the outer electron concentration, s + d of transition metals and the moment of the magnetic Mn atoms P_c. The magnetic moment increases with increasing outer electron. It has been pointed out that the electron/atom ratio for the forming ability of quasicrystalline alloys is limited to very narrow range[27]. Therefore, it is considered that the excess electrons contribute to form the localized magnetic moment, being consistent with the concept of the Pauling valence[28].

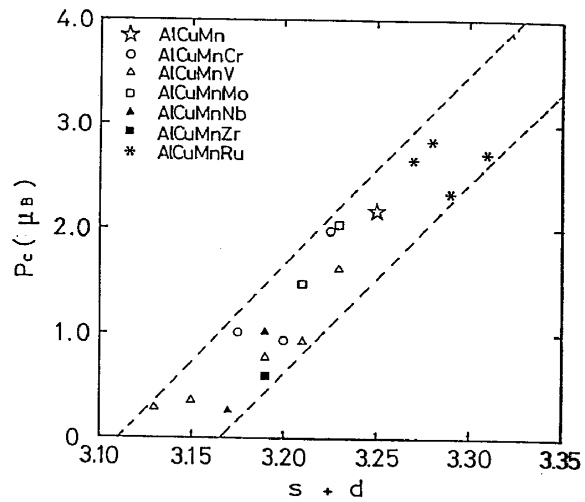

Fig.11 Relationship between Pc and and (s+d) of $Al_{65}Cu_{20}Mn_{15-x}TM_x$.

E) Spin Glass Behavior

From the ratio of the magnetic Mn atoms, the present alloys are regarded as magnetically dilute alloys. The spin glass behavior is often observed in such dilute alloys. Actually, AlMn quasicrystalline

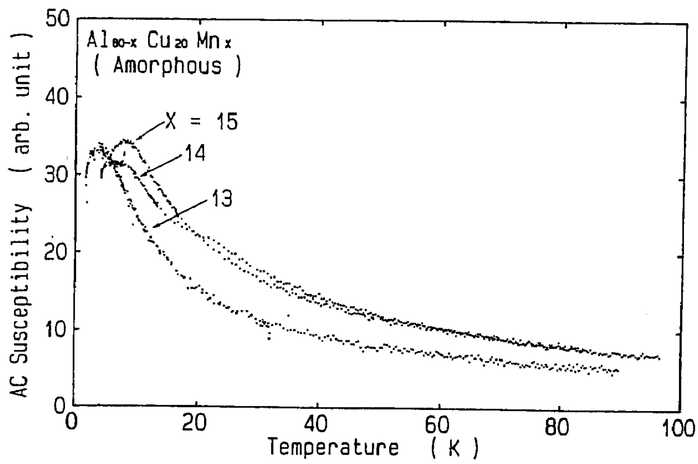

Fig.12 Temperature dependence of AC susceptibility of $A\text{-}Al_{80-x}Cu_{20}Mn_x$ measured in 10 Oe with 80 Hz.

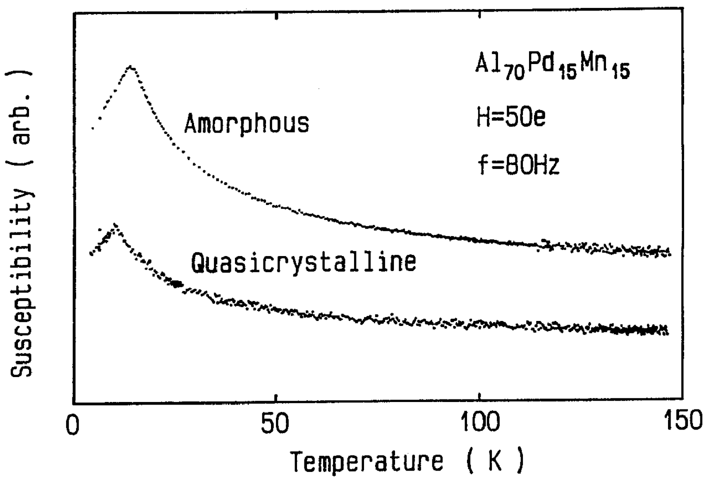

Fig.13 Temperature dependence of Q- and A-$Al_{70}Pd_{15}Mn_{15}$ alloys measured in 5 Oe with 80 Hz.

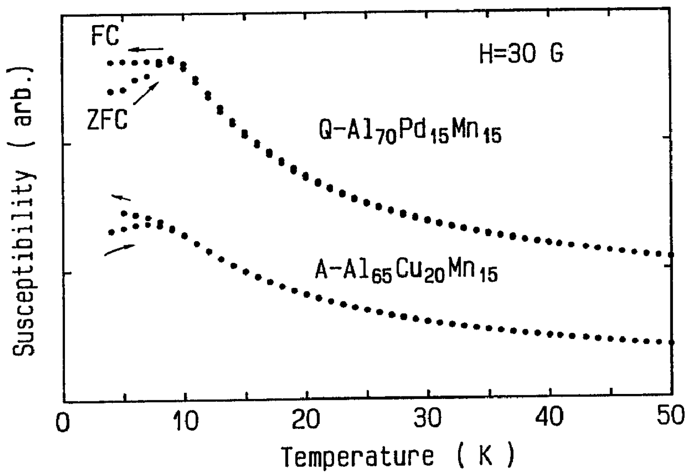

Fig.14 Magnetic cooling effect on susceptibility of Q-$Al_{70}Pd_{15}Mn_{15}$ and A-$Al_{65}Cu_{20}Mn_{15}$ alloys.

and amorphous alloys exhibit the spin glass behavior[3,4]. Figures 12 and 13 show the temperature dependence of AC susceptibility of AlCuMn amorphous and AlPdMn quasicrystalline and amorphous alloys. These alloys exhibit a characteristic cusp of the spin glass. Figure 14 shows the magnetic cooling effect of the representative alloys. They show the clear magnetic cooling effect, resulting in the hysteresis between the zero field cooling(ZFC) and the field cooling in 30 G(FC) at low temperatures. From these measurements, it is concluded that these alloys exhibit a spin glass behavior. Figure 15 shows the concentration dependence of the spin freezing temperature T_f of $Al_{80-x}Cu_{20}Mn_x$ amorphous alloys, $Al_{85-x}Pd_{15}Mn_x$ quasicrystalline and amorphous alloys,

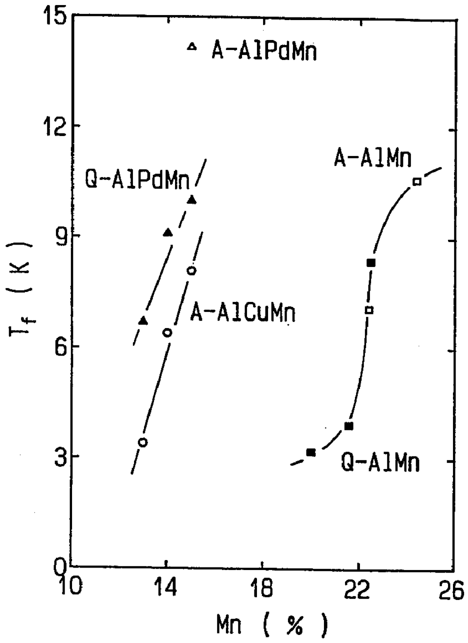

Fig.15 Concentration dependence of the spin freezing temperature of Al-based alloys

together with those of $Al_{100-x}Mn_x$ quasicrystalline and amorphous alloys[3,4]. It is clear that the composition where the spin glass behavior appears depends on the alloy system. Namely, the range of AlCuMn alloys is much lower than that of AlMn alloys and connected with the forming ability of localized magnetic moment and its magnitude. In the present review, many magnetic properties have been presented. In order to make them comprehensible, the formation of localized moment, spin glass behavior and the giant magnetic moment of AlMn, AlCuMn and AlPdMn quasicrystalline and amorphous alloys are tabulated in Table 2,

Table 2 Magnetic state of 3d transition metals in host crystalline metals and in Al-based Q- and A-alloys.

Element and alloy	Impurity or additional element				
	Cr	Mn	Fe	Co	Ni
Al cryst.	N	N	N	N	N
Q-AlTM	N	SG	N	N	N
A-AlTM	N	SG	-	-	-
Cu cryst.	Y	Y	Y	Y	N
Q-AlCuTM	N	Y	Δ	N	N
A-AlCuTM	-	SG	-	-	-
Pd cryst.	Y	SG+GM	GM	GM	Y
Q-AlPdTM	N	SG+GM	Δ	N	-
A-AlPdTM	-	SG+GM?	-	-	-

Y: Localized magnetic moment N: No localized magnetic moment
SG: Spin glass state GM: Giant magnetic moment
Δ : Weak temperature dependent susceptibility -: Unexperimented

together with those of crystalline metals. The magnetic state of impurity in Al, Cu and Pd is given in the very dilute concentration range, compared with that in the quasicrystalline(Q) and amorphous (A) alloys. From the table, it is worth noting that Mn atoms in AlPdMn alloys behave just as those in PdMn crystalline dilute alloys, accompanying the giant magnetic moment and the spin glass behavior.

The decreases in the effective magnetic moment P_{eff} and the saturation magnetization M_s above 14 %Mn shown in Figs.4 and 8, respectively, are not relevant to the second phase because the appearance of the decagonal phase adversely increases those values. Structural experiments are required for a detailed discussion because the effective magnetic moment is not necessarily increased with increasing Mn content, depending on the atomic distance and coordination number[4]. The mean magnetic moment of magnetic Mn atoms in the $Al_{70}Pd_{15}Mn_{15}$ amorphous alloy was estimated to be 5.2 μ_B as shown in Table 1. In order to conclude the presence of the giant magnetic moment in this alloy, however, the ambiguity about 10 % in the extrapolation of the saturation magnetization makes other experiments necessary. Neutron scattering and electron paramagnetic resonance would be expected to help in the understanding of the microscopic properties of Mn in AlPdMn quasicrystalline and amorphous alloys.

Summary

It is difficult to find the difference in the magnetic properties of AlMn alloys between the quasicrystalline and amorphous states. On the other hand, we can see the distinct difference in these properties between the quasicrystalline and amorphous states of AlCuMn and AlPdMn alloy systems. The main results are summarized as follows;

1) The effective magnetic moment of AlCuMn quasicrystalline alloys is smaller than that of amorphous counterparts, and its concentration dependence is not monotonic in contrast to AlMn alloys.
2) AlCuMn amorphous alloys exhibit a spin glass behavior, but the quasicrystalline counterparts show no such behavior. On the other hand, AlPdMn alloys in both states exhibit a spin glass behavior.
3) The ratio of magnetic Mn atoms in AlCuMn and AlPdMn quasicrystalline alloys is about 9%, and that in their amorphous counterparts is about twice.

4) AlPdMn alloys have a giant magnetic moment in a similar manner as PdMn crystalline dilute alloys.
5) By partial substitution of Cr for Mn, the magnetic susceptibility of AlCuMn quasicrystalline alloys is enhanced in analogy with AlMn quasicrystalline alloys. On the other hand, the susceptibility of AlPdMn quasicrystalline alloy is no longer enhanced by partial substitution.

Acknowledgments

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