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Magnetic Properties of Al-Si-MTM (Magnetic Transition Metal) Melt Quenched Amorphous Ribbons*

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Synopsis

Al-Si-MTM(Magnetic Transition Metal) amorphous ribbons were prepared from the melts in order to investigate the magnetic properties. These samples have no magnetic moment, except for Al-Si-Mn amorphous alloys, showing no Curie-Weiss type temperature dependence of the magnetic susceptibility. Al-Si-Mn amorphous alloys exhibit a spin-glass like behavior such as a magnetic cooling effect. The very high electrical resistivity of Al-Si-Mn amorphous alloys is correlated to the magnetic and resonant scatterings. No microcrystalline precipitate was confirmed by electron microscope observation.

Introduction

The electrical and magnetic properties of Al-Mn and Al-Si-Mn alloys in the quasicrystalline and amorphous states have been investigated extensively. The magnetic moment of Al-Mn crystalline alloys is relatively large[1] and other alloy systems such as Al-Co and Al-Ni have no moment at the same concentration range[2]. The electrical resistivity of Al-Mn amorphous alloys is much higher than that of other Al-based alloys[3]. Various kinds of Al-Si-MTM(Magnetic Transition Metal) amorphous alloys were prepared and the electrical resis-

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tivity was measured by Bizen et al.[4]. The electrical resistivity of Al-Si-Mn amorphous alloys is also high compared with that of other alloy systems[4]. The main contributions to the electrical resistivity come from the resonance of the Fermi electrons with the d-state of the transition metals and the structural disorder in the amorphous alloys. Another additional effect is a spin disorder scattering from magnetic spins. Therefore, it is important to study the magnetic properties of Al-Si-MTM amorphous alloys. In the present study, the temperature dependence of magnetic susceptibility, the magnetic cooling effect and ac susceptibility have been investigated.

Experimental

Alloying was repeated several times by arc-melting and several kinds of amorphous ribbons were prepared by melt quenching in an argon gas atmosphere. The amorphous state was confirmed by X-ray diffraction and electron microscopy. The temperature dependence of susceptibility was measured by an induction method using a superconducting magnet. The ac susceptibility was measured by a mutual induction method with a frequency of 80 Hz in a magnetic field of 1.0 Oe. The magnetic cooling effect was measured in the strength of 75 Oe.

Results and discussion

Figure 1 shows the magnetization curves measured up to 60 kOe at 4.2 K for $Al_{58}Si_{25}Mn_{17}$, $Al_{60}Si_{25}Mn_{15}$ and $Al_{60}Si_{25}Cr_{15}$ amorphous alloys. The magnitude of magnetization of Al-Si-Mn amorphous alloys strongly depends on the Mn content and $Al_{58}Si_{25}Mn_{17}$ alloy shows a remarkable field dependence. The magnetization of $Al_{60}Si_{25}Cr_{15}$ alloy increases linearly with increasing magnetic field. Other alloy systems containing Fe, Co and Ni also exhibit a linear change in a similar manner as $Al_{60}Si_{25}Cr_{15}$ amorphous alloy. Therefore, it is expected that Al-Si-Mn amorphous alloys have a magnetic moment, and other $Al_{60}Si_{25}MTM_{15}$ (MTM; Fe, Co, Cr and Ni) amorphous alloys have no magnetic moment.

Figure 2 shows the temperature dependence of magnetization, M, of $Al_{60}Si_{25}Mn_{15}$ and $Al_{60}Si_{25}Cr_{15}$ amorphous alloys. The magnetization of these alloys is extremely small, but the former exhibits a ferromagnetic-like behavior. The susceptibility of a paramagnetic materials is given by the following equation;

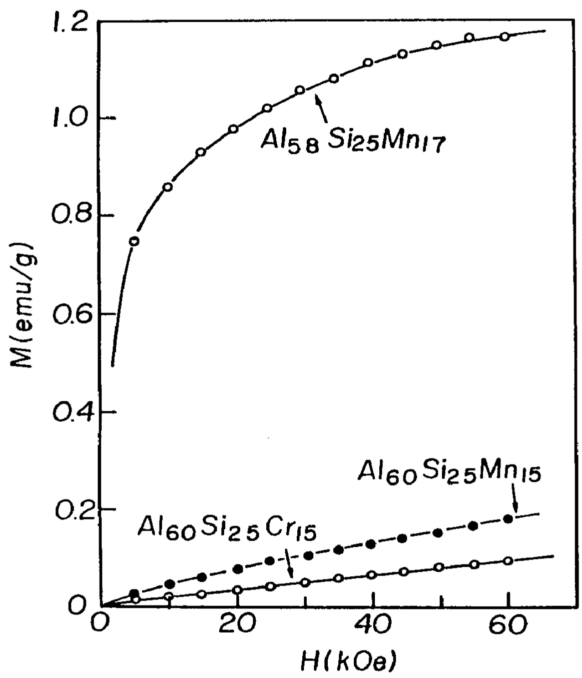

Fig.1 Magnetization curves at 4.2 K for $Al_{58}Si_{25}Mn_{17}$, $Al_{60}Si_{25}Mn_{15}$ and $Al_{60}Si_{25}Cr_{15}$ amorphous alloys.

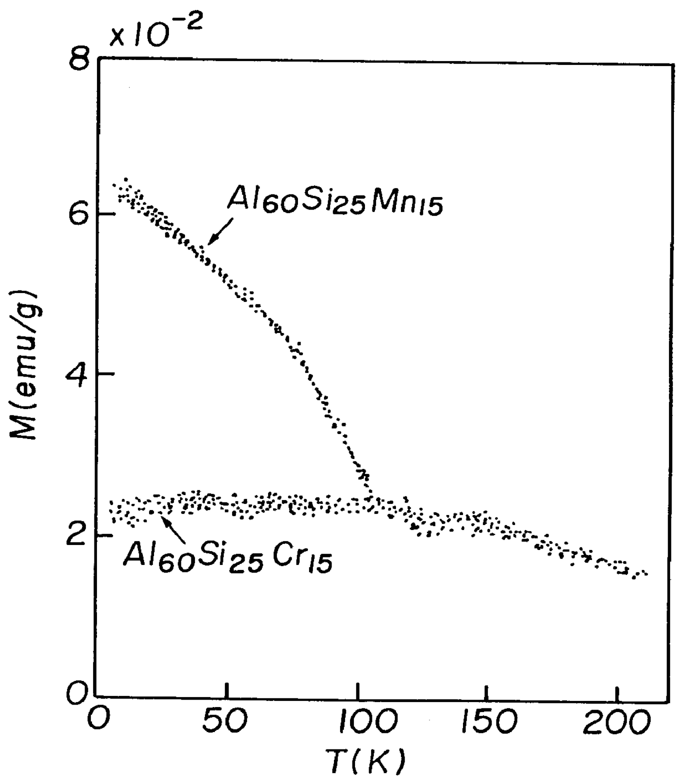

Fig.2 Temperature dependence of magnetization of $Al_{60}Si_{25}Mn_{15}$ and $Al_{60}Si_{25}Cr_{15}$ amorphous alloys.

$$\chi(T) = \chi_{core} + \chi_s(T) + \chi_d(T) \quad (1)$$

with $\chi_s(T) = \chi_s^P(T) + \chi_s^L$ and $\chi_d(T) = \chi_d^P(T) + \chi_d^L + \chi_d^{VV}$, where χ_{core} is the Larmor diamagnetism term, $\chi_{s,d}^L$ and χ_d^{VV} are Landau diamagnetism and Van Vleck paramagnetism terms, respectively. Neglecting the small terms, the total magnetic susceptibility as a function of temperature, $\chi(T)$, is given by the following equation;

$$\chi(T) = \chi_0 - \chi_1 T^2 \quad (2),$$

where χ_0 and $\chi_1 T^2$ are composed of several terms[5]. From this expression, the temperature dependence curve of the susceptibility of paramagnetic materials would become slightly convex upward. The curve of $Al_{60}Si_{25}Cr_{15}$ amorphous alloy would correspond to such a temperature dependence.

The temperature dependence of susceptibility measured in 2 Tesla and its inverse susceptibility of $Al_{58}Si_{25}Mn_{17}$ amorphous alloy is shown in Fig. 3. Above 130 K, a linear slope is obtained, showing a Curie-Weiss type temperature dependence. Mn-metal and Mn-metalloid amorphous alloy systems have been investigated extensively. Ferromagnetic Heusler alloys become spin-glasses due to the frustration of Mn

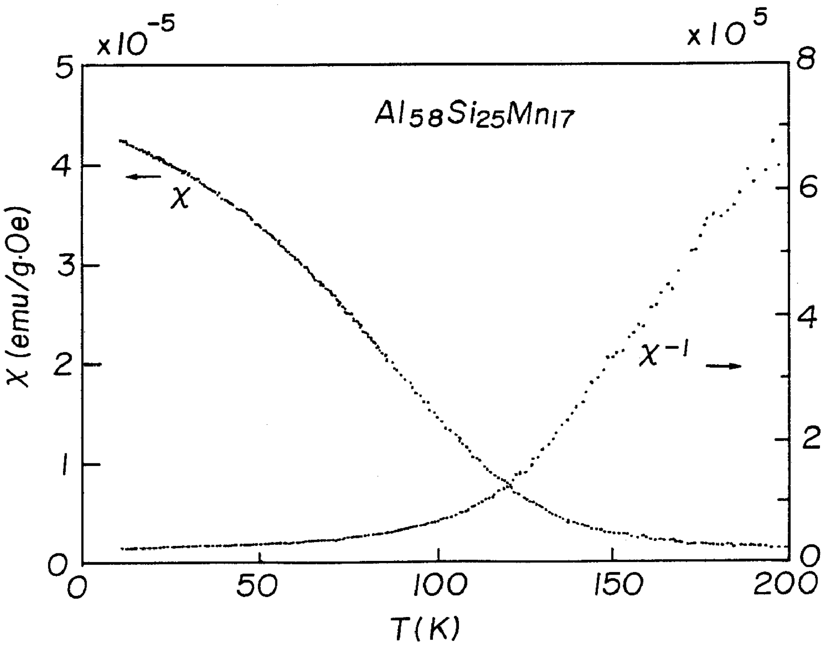

Fig.3 Temperature dependence of the susceptibility and its inverse susceptibility for $Al_{58}Si_{25}Mn_{17}$ amorphous alloy.

moment in the amorphous structure[6]. Hauser et al. have correlated the spin-glass behavior with the d-band mixing[7]. In order to shed light on the ferromagnetic-like behavior of Al-Si-Mn amorphous alloys, ac susceptibility and the magnetic cooling effect have been investigated. Figure 4 shows the temperature dependence of ac susceptibility X_{ac} of $Al_{58}Si_{25}Mn_{17}$ amorphous alloy. The curve exhibits a broad peak at 65 K. The magnetic cooling effect of the same alloy is given in Fig.5. The hysteresis between the zero field cooling(ZFC) and the field cooling(FC) is distinct. Therefore, the magnetic state of this alloy is not a ferromagnetic but a spin glass state. As is well known, the spin glass behavior is often observed very dilute concentration range of Mn. For example, Cu-1%Mn alloy shows a cusp at 12 K[8]. In the case of the present alloy, the Mn concentration is higher than that of Cu-1%Mn by one order of magnitude. Recently, we have measured the high field magnetic properties of Al-Mn quasicrystalline and amorphous alloys and elucidated that only part of Mn atoms carry the magnetic moment[9]. The amorphous structure is very identical with that of the quasicrystalline structure within the second nearest neighbor[10]. Therefore, it is considered that only a fraction of Mn atoms in Al-Si-Mn amorphous alloys has a magnetic moment. Similar ferromagnetic-like behavior has been observed by Hauser et al. in much higher concentration range of Si(30 ∿ 50%) and Mn(20 ∿ 25%)[11]. It is interesting to note that the Curie temperature of those high concentration amorphous alloys is not so different from the apparent Curie temperature obtained from the conventional M^2-T plot for $Al_{60}Si_{25}Mn_{15}$ and $Al_{58}Si_{25}Mn_{17}$ amorphous alloys. Since no microcrystalline precipitates were confirmed by electron microscopy, these magnetic properties are not caused by other phases.

The electrical resistivity of the present amorphous alloys are very high ranging from about 300 ∿ 700 μΩ·cm. Such a high electrical resistivity may be explained by

$$\rho_{total} = \rho_{res} + \rho_{dis} + \rho_{mag} \qquad (3).$$

The high electrical resistivity of amorphous alloys is attributed to the short mean-free-path due to the non-periodicity in the amorphous structure. However, the contribution from only the non-periodicity would not result in such a high electrical resistivity. Because Al-33.3 %Y amorphous alloy exhibits the value of $\rho \sim 150$ μΩ·cm[12], which is very common in amorphous alloys, other contributions such as the resonant state scattering and the disorder scattering from the localized moment would be superimposed. When Mn atoms are dissolved in

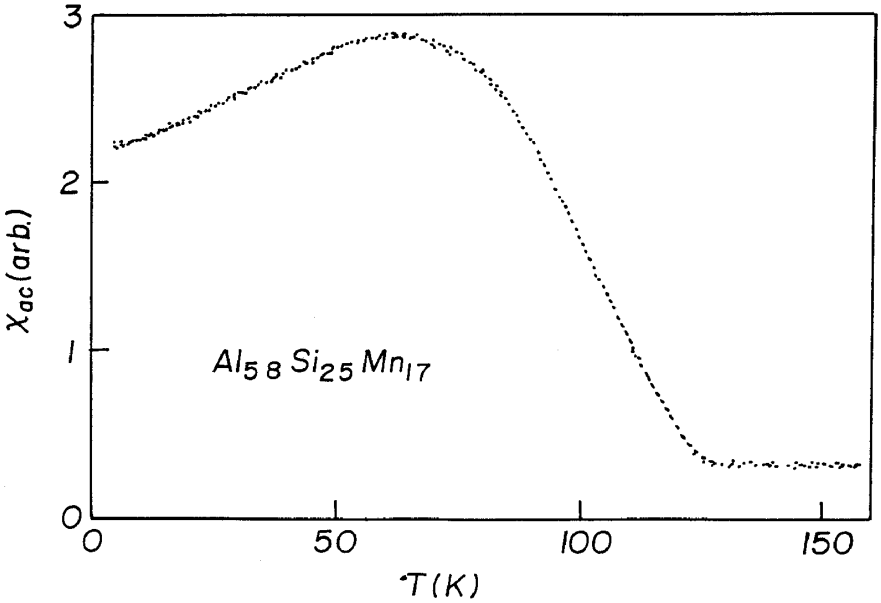

Fig.4 Temperature dependence of ac susceptibility χ_{ac} of $Al_{58}Si_{25}Mn_{17}$ amorphous alloy.

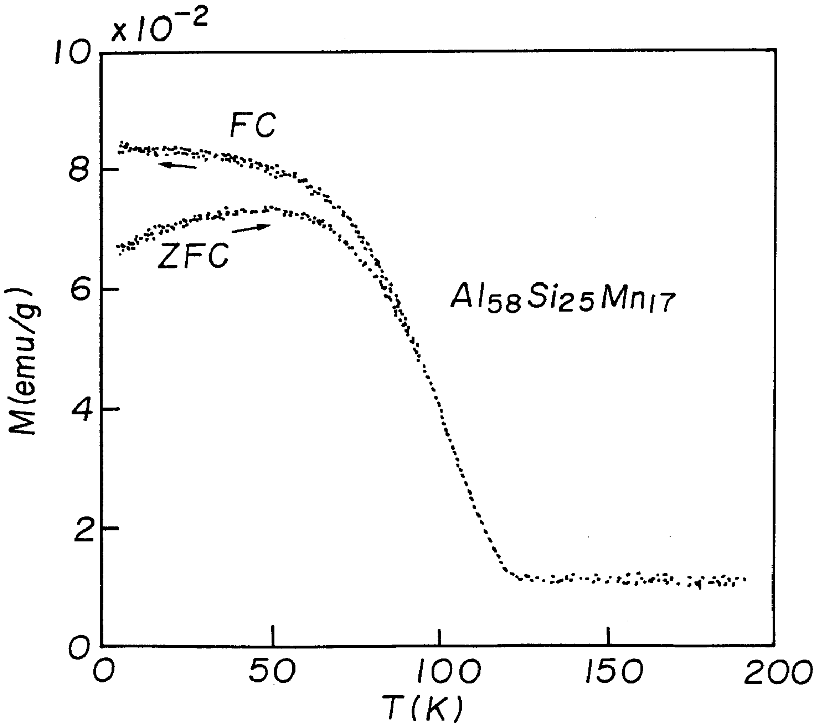

Fig.5 Magnetic cooling effect on the magnetic hysteresis of $Al_{58}Si_{25}Mn_{17}$ amorphous alloy.

Al, a resonant state of the Fermi electrons with the d-state of Mn should take place because the center of the virtual bound state is very close to the Fermi level[13]. The contribution of the resonant state to the electrical resistivity of Al-Mn quasicrystalline alloys has been discussed[14]. In the case of Al-Y alloy system mentioned above, the electrical resistivity is not so high because the Fermi level would not so close to the resonant state. On the other hand, it is worth noting that the electrical resistivity of $Al_{66.7}Gd_{33.3}$ amorphous alloy [12] is much higher by about three times than that of $Al_{66.7}Y_{33.3}$ amorphous alloy due to the additional contribution from the localized magnetic moment. Al-Si-Mn amorphous alloy system has the highest value of electrical resistivity at the same composition among Al-Si-MTM amorphous alloy systems[4]. According to Friedel's virtual bound state model[13], the resonant scattering in Al-Mn is remarkable. Furthermore, Al-Si-Mn amorphous alloys have a localized moment. Therefore, this highest value is explained by taking into account the contribution from ρ_{mag} as well as ρ_{res}.

Summary

Magnetic properties of Al-Si-MTM amorphous ribbons have been investigated. The relationship between the magnetic properties and the electrical resistivity has been discussed. The main results are summarized as follows;

a) Al-Si-Mn amorphous alloys have a magnetic moment, but other alloys have no magnetic moment.
b) The magnetic cooling effect in Al-Si-Mn amorphous alloys is remarkable, showing a spin glass behavior.
c) The temperature dependence curve of susceptibility of $Al_{60}Si_{25}Cr_{15}$ amorphous alloy seems to follow the T^2-low.
d) The magnetic and resonant scatterings are responsible for the highest electrical resistivity of Al-Si-Mn amorphous alloys.

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