Magnetic Properties of Structure-Disordered Heavy Fermion Ce-Mn alloys

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Abstract. We have measured the magnetic susceptibility \( \chi \) on structure-disordered Ce\(_{x}\)Y\(_{80-x}\)Mn\(_{20}\) alloys from 2 to 280 K. The \( \chi \) shows a typical Curie-Weiss (CW) paramagnetic behavior above 20 K in each concentration \( x \). In the Ce-poor region of \( x \leq 51 \), the low-temperature \( \chi \) below 15 K exhibits a spin-glass transition. The spin-glass transition temperature \( T_g \) decreases with increasing Ce-concentration. In the Ce-rich region of \( x \geq 62 \), the spin-glass transition disappears and the CW paramagnetic behavior remains down to the low temperature limit. The Weiss temperature \( \theta \) is negative for all the samples and almost constant in the Ce-poor side. However, the absolute value of \( \theta \) increases with increasing Ce-concentration in the Ce-rich region. We have interpreted this fact as an increase of the antiferromagnetic \( c-f \) coupling, namely the Kondo effect, with the Ce-concentration.

1. Introduction

Coherent-Kondo or heavy fermion state exists in the crystalline Ce-based or U-based compounds with the periodic arrangement of the \( f \)-electron site. On the other hand, the coherent Kondo or heavy fermion properties of several structural disordered Ce-alloys have been reported by Sumiyama et al. [1]. The authors showed a large electronic specific heat coefficient \( \gamma (> 100 \text{ mJ/\text{Ce-molK}}^2) \) and \(-\log T \) dependence in the resistivity of these alloys as an evidence of a heavy fermion state due to antiferromagnetic coupling of the \( 4f \)-electron with conduction electrons (\( c-f \) coupling) at incoherent disordered Ce-sites. Recently, we have reported low temperature properties for structure-disordered (\( a-\))Ce-Mn alloys with wide concentration range [2-4]. The specific heat follows \( \gamma T + \beta T^3 \) at low temperature with a large coefficient \( A \) compared to the case of typical structural-disordered alloys with \( T^2 \) law. Very recently, we have investigated specific heat and resistivity on sputtered \( a-\text{Ce, Y}_{80-\text{Ce}}\cdot\text{Mn}_{20} \) alloys with the substitution of non-magnetic Y for Ce [5]. The \( \gamma \) per Ce is very large and almost independent of the Ce-concentration (\( \gamma_{\text{Ce}} \approx 200 \text{ mJ/\text{Ce-molK}}^2 \)). In the resistivity \( \rho \) for \( x \geq 51 \), the \(-\log T \) dependence has been observed at low temperature, which indicates the formation of the dense Kondo state. The low temperature resistivity for \( x \geq 62 \) exhibits a maximum
Figure 1 shows the temperature dependence of the low-temperature susceptibility $\chi$ for $a$-Ce$_{x}$Y$_{80-x}$Mn$_{20}$ alloys below 50 K. At $T_{\text{max}}$, and then decreases as $\rho \propto A T^2$ in the low temperature limit with a large coefficient $A$ proportional to $(1/T_{\text{max}})^3$, suggesting the formation of coherent-Kondo or heavy fermion state. These experimental results are clear evidence of the formation of the heavy fermion state as the Fermi-liquid ground state even in the structure-disordered system. Furthermore, a recent study of $^{55}$Mn NMR in $a$-Ce$_{62}$Y$_{19}$Mn$_{19}$ by Niki et al. [6] shows that the spin-lattice relaxation rate $1/T_1$ has almost $T$-linear dependence at low temperature, suggesting existence of the density of states of heavy fermion bands on the Fermi surface.

In this study, in order to investigate magnetic properties on the structure-disordered heavy-fermion system, we have measured the magnetic susceptibility for the $a$-Ce$_{x}$Y$_{80-x}$Mn$_{20}$ alloys.

2. Experiments

Amorphous Ce$_{x}$Y$_{80-x}$Mn$_{20}$ alloys were prepared by a dc high-rate sputtering method from the arc melted ingots onto water-cooled Cu substrate. Target materials of Ce$_{x}$Y$_{80-x}$Mn$_{20}$ ($x = 10 - 80$) were made by arc-melting from nominal amount of Ce 99.9 %, Y 99.9 % (NIPPON YTTRIUM Co., Ltd) and Mn 99.9 % (Furuuchi Chemical Corporation), in argon-arc furnace. The chemical compositions of the present alloys were determined by induction-coupled plasma analysis (ICP) as Ce$_{10}$Y$_{70}$Mn$_{20}$, Ce$_{28}$Y$_{52}$Mn$_{20}$, Ce$_{51}$Y$_{30}$Mn$_{19}$, Ce$_{62}$Y$_{19}$Mn$_{19}$, Ce$_{76}$Y$_{9}$Mn$_{15}$ and Ce$_{81}$Mn$_{19}$, respectively. The amorphous structure of sputtered samples was confirmed by an x-ray diffraction analysis (XRD). The magnetic susceptibility has been measured by a conventional SQUID magnetometer in field of 100 Oe from 2 K to 280 K.

3. Results and Discussion

Figure 1 shows the temperature dependence of the low-temperature susceptibility $\chi$ for $a$-Ce$_{x}$Y$_{80-x}$Mn$_{20}$ alloys below 50 K. The $\chi$ for all the samples increases with decreasing temperature above 15 K. With decreasing temperature below 15 K in the Ce-poor region of $x \leq 51$, the $\chi$ exhibits a cusp on the zero-field-cooled susceptibility and separates between the field-cooled and zero-field-cooled susceptibilities below around the maximum temperature as shown in Fig. 1 (a). These behaviors indicate occurrence of a spin-glass (SG) transition in the Ce-poor region due to the magnetic interaction between the Mn magnetic moments as has been reported in $a$-Ce-Mn [2] and $a$-Y-Mn [7] alloys. The transition temperature $T_g$ for $x = 10$ is 12 K, which is almost the same as for $a$-Y$_{80}$Mn$_{20}$ alloy [7]. The $T_g$ decreases with increasing Ce-concentration and becomes 9 K at $x = 51$. In the Ce-rich region of $x \geq 62$, the SG transition disappears and the $\chi$ increases with decreasing temperature down in the low temperature limit as shown in Fig. 1 (b). We have analyzed the obtained experimental data in Curie-Weiss (CW) law.
\[ \chi = \chi_0 + \frac{C}{(T - \theta)} , \tag{1} \]

where \( \chi_0 \) is the temperature-independent term, \( C \) is the Curie constant and \( \theta \) is the Weiss temperature. Figure 2 shows the temperature dependence of the inverse susceptibility where the term \( \chi_0 \) has been subtracted. The \( 1/(\chi - \chi_0) \) for each concentration \( x \) linearly decreases with decreasing temperature in the high-temperature region above 20 K following the CW-law. Figure 3 shows the Ce-concentration dependence of the \( \chi_0 \) for \( a-Ce_xY_{80-x}Mn_{20} \) alloys. The \( \chi_0 \) for the SG-phase of \( x \leq 51 \) is almost constant at about \( 1.0 \times 10^{-6} \) emu/gOe. However, in the Ce-rich region of \( x \geq 62 \) where the SG transition disappears, the \( \chi_0 \) increases rapidly with increasing the Ce-concentration and becomes \( 3.3 \times 10^{-6} \) emu/gOe for \( a-Ce_{81}Mn_{19} \). Figure 4 shows the Ce-concentration dependence of the effective paramagnetic moment \( p_{\text{eff}} \) estimated from the Curie constant \( C \). In the present alloys case, it is considered that the magnetic moment exists in both Mn-ion and Ce-ion. Therefore, the \( C \) in the present data is obtained as

\[ C = \left( N_{Ce} (p_{\text{eff}}[Ce])^2 + N_{Mn} (p_{\text{eff}}[Mn])^2 \right)/3k_B , \tag{2} \]

where \( N_{Ce} \) and \( N_{Mn} \) are a number of Ce-atom and Mn-atom per gram, respectively. However, we cannot estimate separately the \( p_{\text{eff}}[Ce] \) per Ce and \( p_{\text{eff}}[Mn] \) per Mn. So, we have estimated \( p_{\text{eff}} \) as an average value from \( C = (N_{Ce} + N_{Mn})p_{\text{eff}}^2/3k_B \). The average value \( p_{\text{eff}} \) of the \( p_{\text{eff}}[Ce] \) and \( p_{\text{eff}}[Mn] \) is almost constant (\( \approx 1.0 \) \( \mu_B \)) for \( x \leq 76 \). However, the \( p_{\text{eff}} \) for \( a-Ce_{81}Mn_{19} \) becomes 2.0 \( \mu_B \) two times larger than for \( x \leq 76 \). Figure 5 shows the Ce-concentration dependence of the Weiss temperature \( \theta \) estimated from the CW-law. The \( \theta \) shows a negative value for all the samples and is almost constant (\( \theta \approx -5 \) K) in the SG-phase. However, the absolute value of \( \theta \) in the Ce-rich region of \( x \geq 51 \) increases with increasing Ce-concentration. This means an enhancement of antiferromagnetic coupling in \( a-Ce_xY_{80-x}Mn_{20} \) alloys with increasing Ce-concentration with the absence of the SG transition.

We found the \(-\log T\) dependence in the resistivity for \( x \geq 51 \) as an evidence of the dense Kondo state [5]. Therefore, the negative increase of \( \theta \) for \( x \geq 51 \) would arise from the c-f mixing or the antiferromagnetic c-f coupling at each Ce-site, namely the dense Kondo effect. According to the Kondo picture, the \( \theta \) is one of measures for the Kondo temperature \( T_K \) as \( \theta = 2T_K \) [8]. Therefore, we
can conclude that the $T_K$ of $\alpha$-Ce$_x$Y$_{80-x}$Mn$_{20}$ increases with the Ce-concentration for $x \geq 51$. In the present disordered Ce-alloys in the Ce-rich region of $x \geq 62$, coherent-Kondo or heavy fermion state is suggested to form due to the $c$-$f$ mixing or antiferromagnetic coupling between 4$f$-electron of Ce and conduction electrons from the resistivity as shown in Ref. [5]. It has been further pointed out that the temperature $T_{\text{max}}$ at the resistivity maximum below the -log$T$ dependent temperature region is one of scaling parameters of the HF state for the disordered system. The inset of Fig. 5 shows the absolute Weiss temperature $\theta$ vs. the $T_{\text{max}}$ from the resistivity in the HF-phase. We found a linear relation between $\theta$ and $T_{\text{max}}$ as $\theta = \theta_0 + 1.9 T_{\text{max}}$, where $\theta_0$ is 12.7 K. The first term would be interpreted as the local Kondo and/or a conventional AF-interactions and the second term would be interpreted as the coherent effect in the present disordered system. This fact would suggest that the $\theta$ includes an important parameter associated with not only the local Kondo effect with $T_K$ but also the initial formation of the HF state in the disordered system.

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