DENSE KONDO-LIKE BEHAVIOR IN ELECTRIC RESISTIVITY OF AMORPHOUS $Mn_{100-x}Ce_x$ ALLOY*

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Binary amorphous alloy system $Mn_{100-x}Ce_x$ has been fabricated by a dc-sputtering deposition with various Ce compositions ranging from $x = 20$ to 80. Temperature dependence of electrical resistivity for Ce-rich side of amorphous $Mn_{100-x}Ce_x$ alloy, especially in $Mn_{21}Ce_{79}$, exhibits a large initial increase with T^2-like behavior at low temperature side up to about 7K followed by a quite rapid logarithmic-like decrease with the increase of temperature. In contrast, amorphous $Mn_{75}Ce_{25}$ alloy shows only monotonic decrease in the resistivity with increasing temperature. These results reveal a clear evidence of a dense Kondo behavior for Ce rich side.

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1. Introduction

In recent several decades many exciting phenomena have been found for the dense Kondo materials including mainly Ce or Yb and U as one of elements. The $4f$ electrons of Ce ion or $5f$ electrons of U ion have a exchange interaction with the conduction electrons (s, p or d) through the Kondo effect and behave as a itinerantly at low temperature. The characteristic feature of the dense Kondo system appears in large values of linear specific heat coefficient (γ), Pauli paramagnetic susceptibility (χ) and T square coefficient (A) of electrical resistivity.

Amorphous Mn–Ce (a-Mn–Ce) alloy is regarded as one of the candidates belonging to the dense Kondo materials because of having large amount of Ce atoms. From our early study of a-Mn–RE alloys, where RE shows La, Y or

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Sc, these amorphous alloys have been found to show the spin glass characteristics at low temperature. The nature of spin glass characteristics of a-Mn–Y has been investigated theoretically by Kakehashi and Yu(KY) [1] on the basis of the itinerant electron model. Our result of a-Mn–Y alloy [2] shows that the spin–glass ordering temperature T_g depends on linearly against the Mn concentration, suggesting that Y element might not influence the spin system. This is reasonable because Y has no f-electrons and, therefore, no local moments. In contrast, Ce ion of a-Mn–Ce has at most one f-electron. At the high temperature region f-electrons have a localized spins and may interact each other through the RKKY interaction. But in the low temperature region f-electrons are possible to interact to conduction electrons through the Kondo effect. As a result, the spin system of itinerant d-electrons is probably disturbed.

In the present study we have investigated magnetic and electrical properties in order to check for the possibility of the dense Kondo behavior in a-Mn–Ce system by means of the electrical resistance and magnetic susceptibility measurements.

2. Experimental

Amorphous $Mn_{100-x}Ce_x$ alloys (nominally $x = 20, 50, 80$) were prepared by a dc high rate sputtering onto water cooled Cu substrate. The amorphus structure of sputtered samples was confirmed by an X ray diffraction analysis. The chemically analyzed composition is written, for instance $Mn_{75}Ce_{25}$, hereafter. Temperature dependence of the magnetization has been measured by a SQUID magnetometer. The electrical resistance has been measured by a 4 terminal method from about 2 K to 280 K.

3. Results and discussion

The magnetic susceptibility of present a-Mn–Ce alloys shows steep increase with decreasing temperature at experimentally reached lowest temperature limit (about 1.8 K) and thus shows no ordering except for the small kink at low temperature side for $x = 25$ and 50. Usually in Ce based compounds such as Ce–La–Al [3] exhibiting the dense Kondo behavior, the magnetic susceptibility at low temperature region exhibits Pauli paramagnetism resulting in losing the localized moment of Ce atoms. In a-$Mn_{21}Ce_{79}$ the magnetic susceptibility behaves as $\chi \sim 1/(T - \Theta)$ like a Curie–Weiss law down to 1.8 K. This means that the alloy still has individual local moment and the local moment does not order down to low temperature limit. According to KY, Mn's d-electron in a-Mn–Y alloy behaves itinerantly. Average local moments distribute around 0, but Mn spins take an antiferromagnetic coupling. This negative magnetic coupling causes the frustration in the spin system due to the lattice disorder. As a result spin system freezes randomly

at low temperature. Different from a-Mn–Y, Mn spins or even the Ce spins especially in a-$Mn_{21}Ce_{79}$ might not order down to about 1.8 K. This is probably due to the itinerant behavior of Ce $4f$-electrons which prevent the Mn spins from ordering.

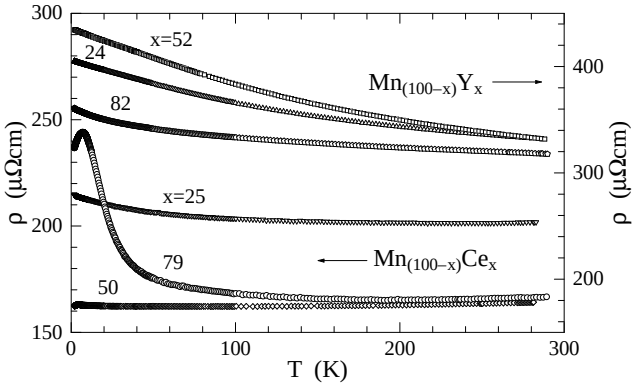

Fig. 1. Temperature dependence of electric resistivity of a-$Mn_{(100-x)}Ce_x$ alloys ($x = 25$, 50 and 79) and a-$Mn_{(100-x)}Y_x$ alloys ($x = 24$, 52 and 82).

The electrical resistivity of a-Mn–Ce are shown in Fig. 1 and that of a-Mn–Y are also shown for comparison. The most striking result is a large increase of resistivity below about 40 K in a-$Mn_{21}Ce_{79}$ with decreasing temperature. Usually amorphous material has a large residual resistivity ρ_0 (more than $100\mu\Omega$cm), mainly due to the random configuration of atoms, and the temperature dependence of resistivity is small with at most 20% change. Referred to as Mooij's empirical law [4], when a residual resistivity exceeds about $200\mu\Omega$cm, the temperature coefficient of resistivity (TCR) becomes negative. All the curves in a-Mn–Y alloy have negative temperature coefficient for all temperature range (2 to 280 K) probably due to the large residual resistivity. ρ_0 of a-$Mn_{21}Ce_{79}$ is about $230\mu\Omega$cm. The resistivity–temperature curve also has a negative TCR as a whole but it shows very steep change below about 40 K. It is rather anomalous. $\rho - \log T$ plot of a-$Mn_{21}Ce_{79}$ is shown in Fig. 2. As seen in Fig. 2, in the temperature range $T \sim$15 to 30 K the logarithmic dependence ($-\log T$) appears clearly. Sometimes amorphous alloys show $\rho - \log T$ dependence. But present steep logarithmic increase of resistivity with decreasing temperature is considered to be the evidence of the Kondo effect. a-$Mn_{75}Ce_{25}$ and a-$Mn_{50}Ce_{50}$ do not show such a behavior. Moreover, the $\rho - T$ of a-$Mn_{21}Ce_{79}$ saturate at around $T \sim 8$ K and then decrease with decreasing temperature. For very low temperature region, it is considered that $\rho - T$ obeys T^2 dependence. In order to determine this behavior accurately, we must erase the other contribution from the net electrical resistivity and pick up the mag-

netic contribution only. ρ_{mag} is connected to the coefficient A as $\rho_{\mathrm{mag}} = \rho_0 + AT^2$. This term is written as $\rho_{\mathrm{mag}} = \rho_{\mathrm{net}} - \rho_{\mathrm{phonon}} - \rho_{\mathrm{electron}} - \rho_{\mathrm{amorphous}}$. Roughly we can put that $\rho_{\mathrm{amorphous}}$ is temperature independent (so we neglect this term, hereafter), and $\rho_{\mathrm{phonon+electron}}$ $(\mathrm{Mn_{21}Ce_{79}}) \sim \rho_{\mathrm{phonon+electron}}$ $(\mathrm{Mn_{18}Y_{82}}) \sim \rho_{\mathrm{net}}$ $(\mathrm{Mn_{18}Y_{82}})$. The difference in ρ_0 between a-$\mathrm{Mn_{21}Ce_{79}}$ and a-$\mathrm{Mn_{18}Y_{82}}$ is considered to be attributed to the difference in $\rho_{\mathrm{amorphous}}$. Thus we calculate the magnetic term of a-$\mathrm{Mn_{21}Ce_{79}}$ as $\rho_{\mathrm{mag}}(\mathrm{Mn_{21}Ce_{79}}) = \rho_{\mathrm{net}}$ $(\mathrm{Mn_{21}Ce_{79}}) - \rho_{\mathrm{net}}(\mathrm{Mn_{18}Y_{82}})$. The result is shown in Fig. 3 as a function of T^2. In this figure, the large negative extrapolated value at zero temperature is attributed to the difference in $\rho_{\mathrm{amorphous}}$. At very low temperature region ρ_{mag} is roughly linear against T^2. The coefficient A is thus estimated to be $\mathrm{A} = 0.75\mu\Omega\mathrm{cmK}^{-2}$. Using γ value referred below we can calculate A/γ^2 as $1.1 \times 10^{-5}\mu\Omega\mathrm{cmK}^{-2}/(\mathrm{mJK}^{-2}\mathrm{mol}^{-1})^2$. This value is quite reasonable compared to other dense Kondo alloys.

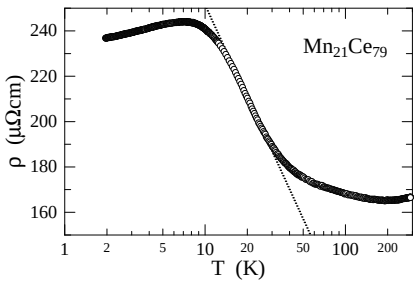

Fig. 2. ρ *vs.* $\log T$ of a-$\mathrm{Mn_{21}Ce_{79}}$ alloy.

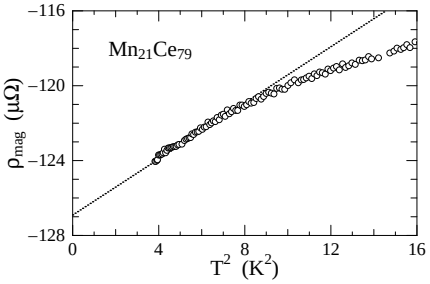

Fig. 3. ρ_{mag} *vs.* T^2 of a-$\mathrm{Mn_{21}Ce_{79}}$ alloy.

The dense Kondo-like behavior is also supported by the large γ value of the specific heat [5] as $\gamma(\mathrm{Mn_{21}Ce_{79}}) \sim 260\,\mathrm{mJ/molK}^2$. Usually amorphous alloy has γ value at most $20\,\mathrm{mJ/molK}^2$ and even in amorphous spin–glass alloy it has at most several decades. So the large value in a-$\mathrm{Mn_{21}Ce_{79}}$ is rather anomalous and we must take the dense Kondo behavior into consideration in order to better understanding this large value.

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