

# Magnetic Properties of Amorphous $Mn_{100-x}Ce_x$ Alloys

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## Abstract

We have measured the magnetization and thermal expansion coefficient  $\alpha$  on sputtered amorphous  $Mn_{100-x}Ce_x$  alloys in various concentrations of Ce. In Mn-rich samples we observe a small anomaly and thermal hysteresis between field-cooling and zero-field-cooling in the low-temperature magnetization, suggesting a spin-glass ordering, and an anomalously large  $\alpha$  value with normal Debye-like temperature dependence. In Ce-rich samples we observe typical Curie-Weiss behavior in the magnetization, but  $\alpha$  increases with the decrease of temperature below 100K, suggesting 4f-electron contribution of the Ce atom.

*Key words:* magnetization; thermal expansion; amorphous alloys; Mn-Ce

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Magnetic properties of Mn-based rare-earth (RE) binary amorphous alloys have not been much investigated in comparison with those of other 3d magnetic element- (such as Fe-, Co- and Ni-) based RE amorphous alloys. Recently, amorphous (*a*-)Mn-Y alloys have been investigated and found to show typical spin-glass properties with the ordering temperature  $T_g$  increasing linearly as a function of Mn concentration [1]. This would be reasonable because Y is assumed to be non-magnetic and Mn is only a magnetic element in this disordered system. Kakehashi and Yu have investigated the nature of the spin glass in *a*-Mn-Y theoretically on the basis of the itinerant electron model [2]. In this study, we replace non-magnetic Y atom in *a*-Mn-Y with another RE element Ce with 4f-electron and investigate magnetic and thermal properties in order to see how the Mn-based amorphous system changes its magnetic characters and how the 4f-electron behaves.

Bulk ingots of  $Mn_{100-x}Ce_x$ , nominally  $x=20,30,40, 60,70$  and 80, were made by melting stoichiometric amount of Mn 99.9% and Ce 99.9% in an argon-arc fur-

nace. Amorphous  $Mn_{100-x}Ce_x$  alloys were prepared by a dc high-rate sputtering method from the arc-melted ingots onto water-cooled Cu substrate. The amorphous structure of sputtered samples was confirmed by an X-ray diffraction analysis. Table 1 shows nominal and chemically analyzed compositions of the sample, respectively. Hereafter, the nominal composition will be used for sample identification. The magnetization has been measured by a conventional SQUID magnetometer in field of 1000e from 4.2K to 300K. The thermal expansion measurements have been done by a strain gauge method for rectangular samples with a typical size of  $10\times 4\times 0.4\text{mm}^3$  in a He gas-flow cryostat from 20K to 300K.

Figure 1 shows the temperature dependence of the magnetization for *a*- $Mn_{100-x}Ce_x$  alloys. In higher temperature region above 20K it shows a typical Curie-Weiss behavior for all the samples. With decreasing temperature below 20K the magnetization for Mn-rich samples ( $x<50$ ) exhibits an anomaly and then indicates difference between field-cooled (FC) and zero-field-cooled (ZFC) magnetization. This would suggest a spin glass ordering, as observed more clearly at higher temperature in *a*-Mn-Y alloys for the same Mn concen-

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		Concentration		
Nominal		Analyzed		Notation
Mn	Ce	Mn	Ce	
20	80	21.1	78.9	Mn <sub>20</sub> Ce <sub>80</sub>
30	70	29.1	71.0	Mn <sub>30</sub> Ce <sub>70</sub>
40	60	40.9	59.1	Mn <sub>40</sub> Ce <sub>60</sub>
60	40	50.0	50.0	Mn <sub>60</sub> Ce <sub>40</sub>
70	30	66.8	33.2	Mn <sub>70</sub> Ce <sub>30</sub>
80	20	75.0	25.0	Mn <sub>80</sub> Ce <sub>20</sub>

Table 1  
Results of quantitative analysis for Mn<sub>100-x</sub>Ce<sub>x</sub> samples by optical emission spectrochemical analysis

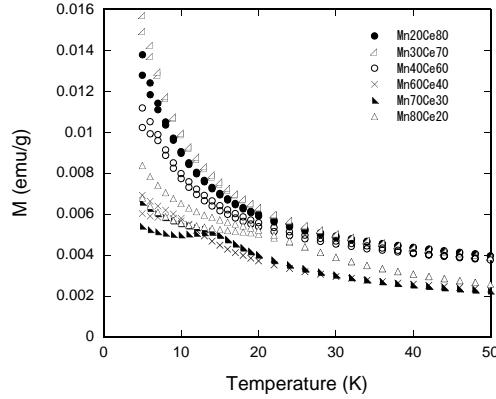


Fig. 1. Temperature dependence of the FC and ZFC magnetization of  $a\text{-Mn}_{100-x}\text{Ce}_x$  alloys in field 100Oe.

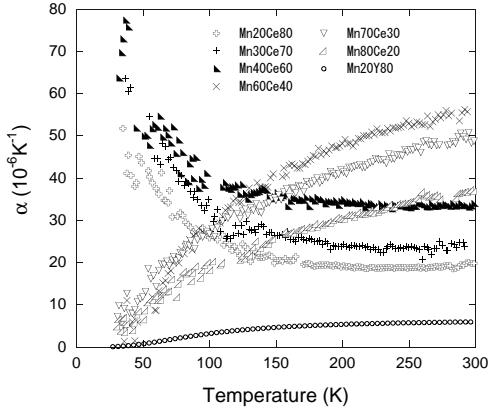


Fig. 2. Temperature dependence of the thermal expansion coefficient  $\alpha$  of  $a\text{-Mn}_{100-x}\text{Ce}_x$  alloys

tration. With increasing Ce concentration the anomaly becomes smaller with a decrease in the temperature of the anomaly and finally disappears in Ce-rich samples ( $x>50$ ), though very small difference between FC and ZFC magnetization remains observable. Therefore, the Ce atom plays a role of reducing Mn-Mn spin interactions in this  $a\text{-Mn-RE}$  system compared to the Y case.

The thermal expansion coefficient  $\alpha$  of  $a\text{-Mn}_{100-x}\text{Ce}_x$

is shown in Fig. 2. For Mn-rich samples ( $x<50$ ) the temperature dependence of  $\alpha$  shows a Debye-like curve, however, its value is anomalously large compared to  $a\text{-Mn}_{100-x}\text{Y}_x$  alloys as shown for  $x=80$  in Fig. 2 as an example. The  $\alpha$  increases with increasing Ce concentration above 100K. According to recent specific heat measurements on these samples [3], the data for  $a\text{-Mn}_{50}\text{Y}_{50}$  also show a Debye-like curve and become almost temperature independent at around room temperature with the value close to  $3R$ , indicating negligible magnetic or electronic contribution. But those for  $a\text{-Mn-Ce}$  is much larger than this value and still increasing with increasing temperature around room temperature, suggesting large magnetic contribution. The present results of  $\alpha$  are consistent with the specific heat and suggests that  $a\text{-Mn-Ce}$  are itinerant magnetic alloys with large spin fluctuations. Such spin-fluctuation contribution to  $\alpha$  has already been reported to exist in  $a\text{-Mn-Y}$  for Mn-rich concentration[4]. For Ce-rich samples ( $x>50$ ),  $\alpha$  largely increases with decreasing temperature below 100K as shown in Fig. 2. Although the  $\alpha$  value decreases with increasing Ce concentration, the temperature variation of  $\alpha$  does not depend on  $x$  very much for  $x>50$ . We suppose that this anomalous behavior below 100K is due to 4f-electron contribution of the Ce atom. There should be two possible explanation for such 4f-electron contribution. One is the crystalline-electric-field (CEF) splitting of the 6-fold degenerate level of Ce<sup>3+</sup> ion with local 4f-electron character [5]. The other is the dense Kondo effect and formation of itinerant 4f-electron state, that is, the heavy-fermion state [6]. The recent specific heat measurements show existence of a dense-Kondo-like behavior with a large linear-specific-heat coefficient  $\gamma$  as  $\sim 260$  mJ/molK<sup>2</sup> for  $a\text{-Mn}_{20}\text{Ce}_{80}$ . Quite recently, the resistivity for  $a\text{-Mn}_{20}\text{Ce}_{80}$  has been measured and found to exhibit a large initial increase with  $T^2$ -like behavior at low temperature followed by a logarithmic-like decrease with the increase of temperature[7]. The present anomaly in  $\alpha$  for Ce-rich samples at low temperature would also suggest the dense-Kondo-like effect observed in the specific heat and resistivity measurements on  $a\text{-Mn}_{20}\text{Ce}_{80}$ .

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