Heavy-fermion like behavior of amorphous Ce$_{x}$Mn$_{100-x}$ alloys system studied by $^{55}$Mn NMR

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Abstract. In order to investigate a heavy-fermion like behavior on Ce-rich side of amorphous Ce$_{x}$Mn$_{100-x}$ alloys system, the $^{55}$Mn NMR measurements on the powdered sample of amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$, being diluted with non-magnetic Y, have been carried out in the temperature range from 1.8 to 270 K. A broadened resonance spectrum containing five NQR lines is observed in the measuring temperature region. Temperature dependences of Quadrupole coupling constant ($e^2Qq$) and the line width of $^{55}$Mn NMR are obtained by means of analysis of each spectrum. Temperature dependence of the line width is expressed by the Curie-Weiss law with $T_c = -12.0$ K. The $1/(T_1T)$ is rapidly increased in proportion to inverse square root of temperature with decreasing temperature, but it keeps almost constant below about 6 K, which shows the heavy-fermion state in Ce$_{62}$Y$_{19}$Mn$_{19}$.

1. Introduction
The structure-disordered type (amorphous) Ce-alloys have been pointed out to have a possibility of forming the dense Kondo (heavy fermion) state at low temperatures [1]. The results of the measurements of the specific heat [2] and electrical resistivity [3] on Ce-rich side of sputtered amorphous Ce$_{x}$Mn$_{100-x}$ alloys reveal the characteristic behavior of the dense Kondo state: at low temperatures the specific heat follows $\gamma T$ with the very large electric specific heat coefficient $\gamma$, and the electrical resistivity depends on $T^2$ law with its large coefficient $A$ in comparison with a typical value for amorphous alloys. The $\gamma$ and $A$ show systematic changes as a function of Ce concentration and the ratio of $A/\gamma^2$ is almost constant. However, it is much smaller than the values in typical heavy fermion systems indicated by the Kadowaki-Woods (KW) relation [4].

The $^{55}$Mn NMR measurements (a nuclear spin $I = 5/2$) on powdered sample of amorphous Ce$_{59}$Mn$_{41}$ have been carried out from 4.2 to 270 K [5]. A broadened resonance spectrum containing five NQR lines has been observed in the measuring temperature region. The temperature dependence of the line width of $^{55}$Mn NMR has been found to be expressed by the Curie-Weiss law with $T_c = -10.5$ K. The nuclear spin-lattice relaxation time $T_1$ has been also measured for amorphous Ce$_{59}$Mn$_{41}$ samples. The $1/(T_1T)$ increases rapidly in proportion to the Curie-Weiss law ($T_c = -0.4$ K) with decreasing temperature, whereas it keeps almost constant below about 10 K, which clearly shows the heavy-fermion like behavior [6].
Figure 1. Powder-pattern spectrum of $^{55}$Mn NMR in amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ at 270 K. The spectrum on the upper part of the figure obtained from NMR measurement. Circles and a solid curve show experimental data and a calculated curve, respectively. The powder-pattern spectrum on the lower part of the figure shows the one obtained in the case that each NQR line in the upper part one has very narrow width.

It is difficult to remove the influence of Mn for the Ce concentration dependence of the specific heat and resistivity in amorphous Ce$_x$Mn$_{100-x}$ because the concentration of Mn besides Ce varies simultaneously with changing $x$. Therefore, the measurements of the specific heat and electrical resistivity in amorphous Ce$_x$Y$_{80-x}$Mn$_{20}$ alloys, in which the amount of Mn is fixed and Ce concentration is diluted with nonmagnetic Y, have been performed in order to clarify the effect of Ce concentration on the dense Kondo state for the amorphous Ce$_x$Mn$_{100-x}$ alloys system [7]. From these results the Ce$_x$Y$_{80-x}$Mn$_{20}$ alloys have been found to indicate the characteristic behavior of the heavy fermion in the Ce-rich region. In this work, the $^{55}$Mn NMR measurements have been performed to investigate the heavy fermion state in amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ from the microscopic point of view.

2. Experimental
Bulk ingots of Ce$_{80-x}$Y$_x$Mn$_{20}$ in Ce-rich side have been made by melting nominal amount of Ce 99.9 %, Y 99.9 % and Mn 99.9 % in an argon-arc furnace. Amorphous Ce$_{80-x}$Y$_x$Mn$_{20}$ have been prepared by a DC high-rate sputtering method from the arc melted ingots onto water-cooled Cu substrate. The amorphous structure has been confirmed by X-ray diffraction analysis. After removal of the copper substrate by mechanical polishing, the chemical compositions of these alloys were determined by induction-coupled plasma analysis (ICP) as Ce$_{62}$Y$_{19}$Mn$_{19}$. The NMR measurements of $^{55}$Mn nuclei on powdered sample of amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ have been carried out from 1.8 to 270 K. $^{55}$Mn NMR spectra have been measured by magnetic field sweep at resonance frequency $f_0 = 79.9$ MHz by means of pulse sequence $90^\circ$ -t$_1$-90$^\circ$-t$_2$-180$^\circ$ with a conventional pulse spectrometer.

3. Results and Discussion
In general, the spectrum of $^{55}$Mn NMR is split into five resonance lines due to the influence of NQR interaction because of $I = 5/2$. However, in amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ as shown on
Figure 2. Temperature dependence of line width of $^{55}\text{Mn}$ NMR in amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$. The solid curve is obtained by the Curie-Weiss law with $T_c = -12.0$ K.

the upper part of Fig. 1, only single broadened resonance spectrum of $^{55}\text{Mn}$ NMR has been obtained at each temperature between 1.8 and 270 K. This means that each NQR resonance line is broadened owing to the magnetic influences and the broadened resonance spectrum contains all of five NQR lines. The spectrum of amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ is much broadened compared with that of amorphous Ce$_{59}$Mn$_{41}$. Circles in Fig. 1 show the echo intensities obtained from $^{55}\text{Mn}$ NMR measurement. A solid curve shows the calculated powder-pattern spectrum consisting of five NQR lines obtained when asymmetry parameters $\eta$ is 0 and each of five NQR lines consists of Lorentzian one with the same width. Resonance lines of Lorentzian and Gaussian type for each NQR line have been taken for the fitting procedure. Lorentzian lines have been well fitted for the spectra in the measuring temperature region as compared with Gaussian ones. The spectrum in the lower part of Fig. 1 shows the powder-pattern spectrum obtained in the case that each NQR line in the upper part one has very narrow width. Quadrupole coupling constant $\nu_Q$, which is given as $3e^2Qq/(2I(2I-1))h$, is gradually changed from about 0.9 MHz at 1.8 K to about 0.8 MHz at 270 K.

The temperature dependence of the line width of $^{55}\text{Mn}$ is shown in Fig. 2, which can be expressed by the Curie-Weiss law with $T_c = -12.0$ K. The line width shown in Fig. 2 represents that of each Lorentzian-line given for five NQR lines. The temperature dependence of magnetic susceptibility of amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ is also expressed by the Curie-Weiss law [8]. The Curie temperature obtained from the susceptibility is almost consistent with that of NMR result. The origin of temperature variations for both of line width and magnetic susceptibility showing the Curie-Weiss law should be considered to be the same. We could not find the heavy fermion-like behavior from the temperature dependence of the NMR line-width in the low temperature region. As line width as well as magnetic susceptibility is sensitive to static magnetic states and spatial magnetic randomness in the sample, they would be much affected by these magnetic aspects in amorphous Ce-Mn system.

It has been extremely difficult to determine the values of Knight shift because broadened resonance line becomes much broader due to the Curie-Weiss law with decreasing temperature. However, the values of Knight shift is almost constant within experimental accuracy from 1.8
Figure 3. Temperature dependence of $1/(T_1 T)$ of $^{55}$Mn NMR in amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$. The solid line is in proportion to inverse square root of temperature.

to 270 K.

As shown in Fig. 3, the $1/(T_1 T)$ of $^{55}$Mn NMR in amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ increases rapidly in proportion to inverse square root of temperature with decreasing temperature, whereas it keeps almost constant below about 6 K, which ascribes to the heavy-fermion state. On the other hand, in amorphous Ce$_{59}$Mn$_{41}$, it has been found that the $1/(T_1 T)$ of $^{55}$Mn NMR increases rapidly in proportion to the Curie-Weiss law ($T_C = -0.4$ K) with decreasing temperature and it keeps almost constant below about 10 K [7]. The temperature dependence of the $1/(T_1 T)$ in proportion to inverse square root of temperature in amorphous Ce$_{62}$Y$_{19}$Mn$_{19}$ would be originated from the substitution of Y for Ce. Further investigations by NMR to elucidate the heavy fermion state of amorphous Ce$_{x}$Y$_{80-x}$Mn$_{20}$ are needed and are now in progress.

4. References
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