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NMR STUDY OF R_6Mn_{23} COMPOUNDS

H. Nagai ([1]), K. Kojima ([2]) and H. Yoshie ([1])

([1]) *Department of Physics, Faculty of Science, Shinshu University, Matsumoto 390, Japan*
([2]) *Faculty of Integrated Arts and Science, Hiroshima University, Hiroshima 730, Japan*

Abstract. – The ^{55}Mn NMR spectra have been observed under zero and external field up to 50 kOe. The negative field dependence of the resonance frequency is obtained for all compounds and explained by the weak anisotropy. The transferred hyperfine fields in Dy_6Mn_{23} and Ho_6Mn_{23} are small in magnitude and comparable with those of Y_6Mn_{23}.

The compounds R_6Mn_{23} have a cubic Th_6Mn_{23}-type structure which is most stable in the intermetallic compounds between rare-earth (R) and Mn atoms (RMn_2, R_6Mn_{23} and RMn_{12}). There are crystallographically four inequivalent Mn sites (4b, 24d, $32f_1$ and $32f_2$) with a population ratio of 1;6;8;8. The high magnetic ordering temperatures of R_6Mn_{23} indicate the strong coupling between the magnetic moments, while the Mn moments are estimated to be very small (0.4 μ_B/Mn) by the magnetic measurements. The R and Mn moments are antiparallel to each other except in Gd_6Mn_{23} [1]. Delapalme *et al.* [2] investigated the ferrimagnetic arrangement of Mn moments in Y_6Mn_{23} by the polarized neutron experiments. They estimated the Mn moments to be -2.81 μ_B (4b), -2.0 μ_B (24d), 1.79 μ_B ($32f_1$) and 1.77 μ_B ($32f_2$) at 4.2 K.

The NMR method is useful for the microscopical investigation of the magnetic properties of these compounds. In this paper we report the ^{55}Mn NMR of R_6Mn_{23} (R = Gd, Dy and Ho) and discuss the hyperfine fields of these compounds.

The samples were prepared by melting the constituent elements of 99.9 % in purity in an induction furnace under a purified argon atmosphere. A small amount of excess Mn metal was added to avoid weight loss due to evaporation. The X-ray diffraction shows the single phase of Th_6Mn_{23}-type structure after annealing at 900 °C for a few days.

Figure 1 shows the typical ^{55}Mn NMR spectra of R_6Mn_{23} (R = Gd and Ho) at 4.2 K under zero field. These spectra are mainly due to the nuclei located in domain walls. The field dependences of the ^{55}Mn NMR spectra are plotted in figure 2 where the lines indicate the ^{55}Mn resonances as a function of the external field with the slope of $-^{55}\gamma$ (= 1.05 kHz/Oe, nuclear gyromagnetic ratio).

There exist mainly three peaks in eachy ^{55}Mn spectra due to the nuclei located in magnetic domains and assigned to the different crystallographic sites. They are denoted as A, B and C. Since the intensity of the lowest frequency peak (C) is much stonger than the others, it may be attributed to $32f_1$ and $32f_2$ sites whose moments are almost the same [3, 4]. The peaks

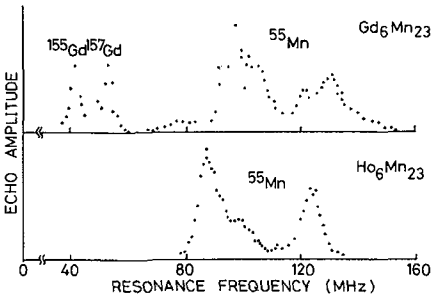

Fig. 1. – ^{55}Mn, ^{155}Gd and ^{157}Gd NMR spectra by the spin-echo method under zero field at 4.2 K. The signals are attributable to the nuclei in domain walls.

A and B may be due to 4b and 24d sites, respectively, on the consideration of their Mn moments. These site assignments are consistent with those of Y_6Mn_{23} by Yoshimura *et al.* [4].

Since the magnetic moments of R atoms couple antiferromagnetically with Mn moments and the magnetic moment of R-sublattice is larger than that of Mn-sublattice, the field dependence of the ^{55}Mn resonance frequency is expected to be in up slope $\left(+^{55}\gamma\right)$ by assuming the hyperfine field at ^{55}Mn nucleus to be negative in sign. It is surprising that the field dependence of the ^{55}Mn resonance frequency is in down slope $\left(-^{55}\gamma\right)$ as shown in figure 2. The negative field dependence may be due to the weak anisotropy in these compounds and ferrimagnetism is easily broken by applying an external field like in Y_6Mn_{23} [3, 4]. The ^{55}Mn NMR spectra under zero field are extremely enhanced by domain wall motions. Then we estimate the values of the hyperfine fields by extrapolating to $H = 0$ in the field dependence of the resonance frequency and the values are listed in table I (in kOe).

The transferred hyperfine fields $\left(H_t^R\right)$ from Dy and Ho are very small for the signals A and C, because the hyperfine fields are almost the same with those of Y_6Mn_{23}. For the signal B the H_t^R is proportional to $-3.1\,(g-1)\,J$ [kOe] for R = Gd, Dy and Ho. In the case of Gd_6Mn_{23}, the hyperfine fields of signals

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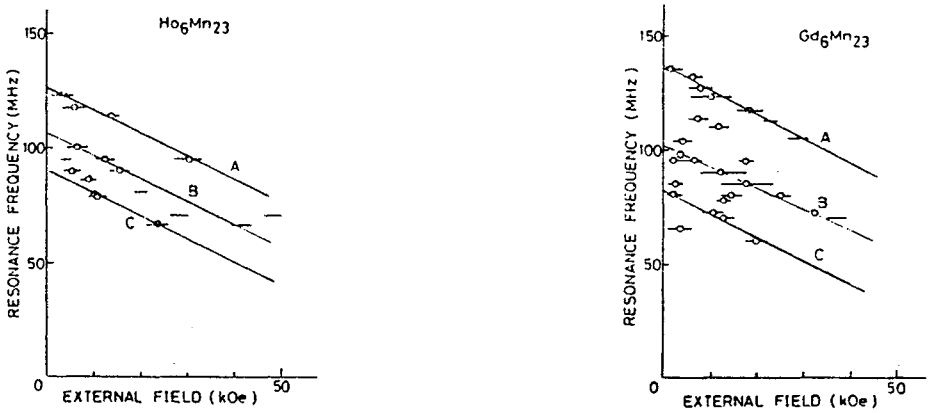

Fig. 2. – The field dependence of ^{55}Mn resonance frequency in Gd_6Mn_{23} and Ho_6Mn_{23} at 4.2 K. The solid and dotted lines are plotted with the slope of $-^{55}\gamma$ values. The lines A, B and C denote the main peaks of signals.

Table I. – *The* ^{55}Mn *hyperfine fields in* R_6Mn_{23} (kOe).

Signal	Site	Gd_6Mn_{23}	Dy_6Mn_{23}	Ho_6Mn_{23}	(Y_6Mn_{23})
A	4b	−131	−122	−122	−122
B	24d	− 98	− 99	−101	−109
C	$32f_1$ } $32f_2$ }	− 79	− 86	− 86	− 88

The data of Y_6Mn_{23} are taken from reference [4].

A and C largely deviate from those of Dy_6Mn_{23} and Ho_6Mn_{23}. Other signals than signals A, B and C were observed as shown in figure 2. These anomalous phenomena are originated from the complex magnetic structure of Gd_6Mn_{23}.

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