Atomic occupancy preference of Ga and Ti and its effect on the Mn spin arrangements in YMn₆Sn₆: Evidence from¹¹⁹Sn Mössbauer spectroscopy

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Abstract

The effects of Ga and Ti substitutions for Sn on the Mn spin arrangements in the YMn₆Sn₆ compound have been investigated by means of X-ray diffraction (XRD), magnetization measurement and Mössbauer spectroscopy. XRD refinement indicates that Ga and Ti atoms prefer to occupy 2d and 2e sites, respectively. Mössbauer spectra indicate incommensurate magnetic structures for the YMn₆Sn₆, YMn₆Sn₅.₄Ga₅.₆ and YMn₆Sn₅.₄Ti₅.₆ compounds. The substitution of Ga for Sn at the 2d sites is found to significantly decrease the turn angle in the Mn–Sn₃–Sn₂–Sn₃–Mn layer and to subsequently increase the net magnetization. On the other hand, the substitution of Ti for Sn at 2e sites increases the turn angles in the Mn–(Y,Sn₁)–Mn and Mn–Sn₃–Sn₂–Sn₃–Mn layers, and consequently decreases the exchange coupling of Mn–Mn between different layers. The resulting ferromagnetic and ferrimagnetic behavior, as has been observed for YMn₆Sn₅.₄Ga₅.₆ and YMn₆Sn₅.₄Ti₅.₆, respectively, can be explained in terms of these spin interactions.

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1. Introduction

Extensive studies have been focused on the magnetic properties of the series of RMn₆X₆ (R=rare earth element and X=Sn or Ge) compounds with the HfFe₆Ge₆-type structure and have demonstrated a wide variety of magnetic behavior [1–4]. The space group of RMn₆Sn₆ compounds is P6/mmm where R atoms occupy 1a(0,0,0) sites, Mn atoms occupy 6i(0.5,0,Zₘₙ=0.25) sites and Sn atoms occupy three non-equivalent sites, 2c(1/3,2/3,0), 2d(1/3,2/3,0.5) and 2e(0,0,0,Zₘₙ=0.34), which are defined as Sn₁, Sn₂ and Sn₃ sites, respectively. These compounds consist of the alternating layers Mn–(R,Sn₁)–Mn and Mn–Sn₃–Sn₂–Sn₃–Mn stacked along the c-axis: the slab. In all of these compounds, the magnetic structure of the Mn atoms in the (001) basal plane is ferromagnetic, while the arrangement of Mn magnetic moments between different layers is complicated and depends on the nature of the R atoms [5–7].

In previous work, we have demonstrated that the substitution of Ga or Ti for Sn results in a significant increase in magnetization, implying a transition from antiferromagnetic coupling to ferromagnetic or ferrimagnetic coupling [8,9]. However, the preferred occupancy of Ga and Ti atoms and its effect on the Mn spin arrangements in different layers of the YMn₆Sn₆ compound is not yet understood in detail. ¹¹⁹Sn Mössbauer spectroscopy is a very efficient tool for probing the local environment of Sn atoms. In magnetically ordered materials, the magnetic hyperfine interaction at the Sn nuclei is related both to the arrangement of the magnetic atoms and to the chemical bonding between Sn atoms and these magnetic atoms. In this work, we present ¹¹⁹Sn Mössbauer spectra in order to investigate the spin arrangements in YMn₆Mn₆, YMn₆Sn₅.₄Ga₅.₆ and YMn₆Sn₅.₄Ti₅.₆. The reasons for using

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Mössbauer spectroscopy are two-fold. Firstly, the large amount of Sn present in these materials provides an intense absorption signal for Mössbauer measurements. Secondly, although all Sn atoms at the three non-equivalent Sn sites have six Mn atoms as the nearest neighbors, the six Mn atoms are located in different layers. Sn1 and Sn2 atoms are located in the Mn–(R,Sn1)–Mn and Mn–Sn3–Sn2–Sn3–Mn layers, respectively, whereas Sn3 atoms have six Mn atoms in the (001) basal plane as the nearest neighbors. Therefore, the intralayer and interlayer spin arrangements of the Mn moments can be observed by their effects on the 119Sn hyperfine fields at the three Sn sites.

2. Experimental methods

Three samples with compositions of YMn6Sn6, YMn6Sn5.4Ga0.6, and YMn6Sn5.4Ti0.6 were prepared by arc melting constituent elements in a highly purified Ar atmosphere. The purity of the elements was at least 99.9%. About 5 and 15 at.% excess of Y and Mn were added to compensate for the mass loss during melting, respectively. The ingots were re-melted at least three times to ensure homogeneity and annealed in an evacuated quartz tube at 1043 K for 240 h then rapidly quenched into water. The crystal structure was confirmed by room temperature (RT) X-ray diffractometer (XRD) scans using CuKα radiation. The crystal structure of the powder samples was determined by Rietveld profile refinement of the X-ray diffraction (XRD) pattern. A superconducting quantum interference device (SQUID) magnetometer was used to measure the magnetic properties of all samples for temperatures below 400 K. 119Sn Mössbauer spectroscopy was used to investigate the hyperfine fields at the three non-equivalent Sn sites. Room temperature 119Sn Mössbauer spectra were recorded with a Wissel System constant acceleration Mössbauer spectrometer with a Ca119mSnO3 source. A palladium foil was used as a critical absorber for Sn X-rays. The Mössbauer spectra were fitted with a least-squares program assuming Lorentzian peaks. The velocity scale was calibrated using the peak-to-peak splitting of the outer lines of Co57Sn ~ 13.143 mm/s and the center shifts are referenced to room temperature CaSnO3.

3. Results and discussions

XRD patterns show that all samples crystallize with the hexagonal HfFe6–Ge6-type structure (P6/mmm space group). Rietveld profile refinement was carried out using the Hunter and Howard package to determine lattice constants and atomic positions [10]. Fig. 1 illustrates the observed and fitted XRD patterns of YMn6Sn6, YMn6Sn5.4Ga0.6, and YMn6Sn5.4Ti0.6 compounds. A few percent of impurity phases were detected in YMn6Sn6 and YMn6Sn5.4Ga0.6 compounds. The crystallographic parameters of the HfFe6–Ge6-type structure (P6/mmm space group) were used to start the refinement. Y and Mn atoms are located at 1a(0,0,0) and 6i(0.5,0,ZMn ~ 0.25) sites, respectively. Initially, it was assumed that Ga(Ti) and Sn atoms occupy the three non-equivalent sites. Room temperature 119Sn Mössbauer spectra were recorded with a Wissel System constant acceleration Mössbauer spectrometer with a Ca119mSnO3 source. A palladium foil was used as a critical absorber for Sn X-rays. The Mössbauer spectra were fitted with a least-squares program assuming Lorentzian peaks. The velocity scale was calibrated using the peak-to-peak splitting of the outer lines of Co57Sn ~ 13.143 mm/s and the center shifts are referenced to room temperature CaSnO3.

Fig. 1. XRD patterns of YMn6Sn6, YMn6Sn5.4Ga0.6, and YMn6Sn5.4Ti0.6. The observed data and fitted patterns are indicated by closed circles and solid lines, respectively. The lowest curves are the difference between observed and calculated patterns. The vertical bars indicated the expected reflection peaks’ positions.
For YMn₆Sn₆, the magnetization increases slowly in applied field of less than 20 and 10 kOe at 5 and 293 K, respectively, implying an antiferromagnetic behavior. In higher applied fields, a field-induced magnetic phase transition was observed in this sample. For the samples of YMn₆Sn₅.₄Ga₀.₆ and YMn₆Sn₅.₄Ti₀.₆, a significant increase in magnetization was achieved in very low magnetic field, suggesting a ferromagnetic or ferrimagnetic behavior. From the magnetization curves, it can be found that the substitution of Ga or Ti leads to an increase in the saturation magnetization. However, the average Mn magnetic moments at 5 K are 1.0, 2.0 and 1.5 μB for YMn₆Sn₆, YMn₆Sn₅.₄Ga₀.₆ and YMn₆Sn₅.₄Ti₀.₆, respectively, which are far smaller than those obtained from neutron diffraction (2.2μB).

Magnetization measurements suggest that a non-collinear magnetic structure exists in these compounds, and that the substitution of Ga or Ti significantly modifies the spin arrangements of Mn atoms.

Although previous ¹¹⁹Sn Mössbauer studies of YMn₆Sn₆ have been reported, the experimental data, taken prior to neutron diffraction experiments, were misinterpreted and the authors concluded an antiferromagnetic arrangement for the Mn moments in the (001) plane [12–14]. A careful ¹¹⁹Sn Mössbauer spectroscopy study of YMn₆Sn₅.₄Mₓ (M=Ga or Ti) is helpful in determining the evolution of the magnetic structure of these materials. Fig. 4 illustrates ¹¹⁹Sn Mössbauer spectra of YMn₆Sn₆, YMn₆Sn₅.₄Ga₀.₆, and YMn₆Sn₅.₄Ti₀.₆ compounds at room temperature. The existence of three non-equivalent Sn sites in these compounds results in very complicated absorption spectra. The central doublets for YMn₆Sn₆ and YMn₆Sn₅.₄Mₓ are related to the Sn-containing impurity phase, which was also observed from XRD patterns. The magnetically split spectra were fitted with three sextets. The resulting hyperfine parameters are given in Table 2.

The measured hyperfine fields are in good agreement with those reported by Amako et al. [12,13]. The substitution of Ga results in an increase in average hyperfine field, while Ti substitution leads to a decrease of average hyperfine field due to the change of the saturation magnetization and Curie temperature.

### Table 1

| Atom site          | Occupancy | x   | y   | z   |
|--------------------|-----------|-----|-----|-----|
| YMn₆Sn₆ P6/mmm a = b = 5.5320(3) Å, c = 9.0122(6) Å |
| Y(1a)              | 1.00      | 0.0000 | 0.0000 | 0.0000 |
| Mn(6i)             | 1.00      | 0.5000 | 0.0000 | 0.2482(9) |
| Sn₁(2c)            | 1.00      | 0.3333 | 0.6667 | 0.0000 |
| Sn₂(2d)            | 1.00      | 0.3333 | 0.6667 | 0.500 |
| Sn₃(2e)            | 1.00      | 0.0000 | 0.0000 | 0.3336(6) |
| YMn₆Sn₅.₄Ga₀.₆ P6/mmm a = b = 5.5045(2) Å, c = 8.9699(4) Å |
| Y(1a)              | 1.00      | 0.0000 | 0.0000 | 0.0000 |
| Mn(6i)             | 1.00      | 0.5000 | 0.0000 | 0.2482(8) |
| Sn₁(2c)            | 0.94(1)   | 0.3330 | 0.6667 | 0.0000 |
| Sn₂(2d)            | 0.75(1)   | 0.3333 | 0.6667 | 0.500 |
| Sn₃(2e)            | 1.00      | 0.0000 | 0.0000 | 0.3364(6) |
| Ga(2c)             | 0.06(1)   | 0.3333 | 0.6667 | 0.0000 |
| Ga(2d)             | 0.25(1)   | 0.3333 | 0.6667 | 0.500 |
| Ga(2e)             | 0.00      | 0.0000 | 0.0000 | 0.3364(8) |
| YMn₆Sn₅.₄Ti₀.₆ P6/mmm a = b = 5.5557(3) Å, c = 9.0634(7) Å |
| Y(1a)              | 1.00      | 0.0000 | 0.0000 | 0.0000 |
| Mn(6i)             | 1.00      | 0.500  | 0.0000 | 0.2475(9) |
| Sn₁(2c)            | 0.93(1)   | 0.3333 | 0.6667 | 0.0000 |
| Sn₂(2d)            | 0.93(1)   | 0.3333 | 0.6667 | 0.500 |
| Sn₃(2e)            | 0.86(1)   | 0.0000 | 0.0000 | 0.3355(7) |
| Ti(2c)             | 0.07(1)   | 0.3333 | 0.6667 | 0.0000 |
| Ti(2d)             | 0.07(1)   | 0.3333 | 0.6667 | 0.500 |
| Ti(2e)             | 0.14(1)   | 0.0000 | 0.0000 | 0.3355(7) |

Fig. 3(a) and (b), respectively. For YMn₆Sn₆, the magnetization increases slowly in applied field of less than 20 and 10 kOe at 5 and 293 K, respectively, implying an antiferromagnetic behavior. In higher applied fields, a field-induced magnetic phase transition was observed in this sample. For the samples of YMn₆Sn₅.₄Mₓ and YMn₆Sn₅.₄Mₓ, a significant increase in magnetization was achieved in very low magnetic field, suggesting a ferromagnetic or ferrimagnetic behavior. From the magnetization curves, it can be found that the substitution of Ga or Ti leads to an increase in the saturation magnetization. However, the average Mn magnetic moments at 5 K are 1.0, 2.0 and 1.5 μB for YMn₆Sn₆, YMn₆Sn₅.₄Ga₀.₆ and YMn₆Sn₅.₄Ti₀.₆, respectively, which are far smaller than those obtained from neutron diffraction (2.2μB).
In order to assign the three subspectra to the three non-equivalent sites, Sn$_1$, Sn$_2$ and Sn$_3$, an analysis of the atomic environment of these three Sn sites is necessary. On the basis of the crystal structure of YMn$_6$Sn$_6K_xM_x$ (M=Ga or Ti) compounds, the nearest neighbors of the Sn$_1$, Sn$_2$ and Sn$_3$ sites are plotted in Fig. 5. Since, $^{119}$Sn hyperfine fields at the Sn sites result from the presence of Mn magnetic moments, only the nearest neighboring Mn atoms are taken into account in this analysis. Although all Sn atoms at these three non-equivalent sites have six Mn atoms as the nearest neighbors, the distribution of the six surrounding Mn atoms is different. The Sn$_1$ and Sn$_2$ sites are situated in the center of a trigonal Mn prism formed within Mn–(Y,Sn)–Mn and Mn–Sn$_3$–Sn$_2$–Sn$_3$–Mn layers, respectively. The Sn$_3$ atom is slightly outside of the hexagonal Mn plane. Since the Sn atoms are non-magnetic, the hyperfine field ($H_{HF}$) acting on $^{119}$Sn nuclei in the magnetically ordered state of YMn$_6$Sn$_6K_xM_x$, arises from the transferred hyperfine field caused by the neighboring magnetic atoms. This is proportional to the number of nearest neighbor magnetic atoms and their magnetic moments;

$$H_{HF} = \sum_j C_i \mu_j(i)$$

where $\mu_j(i)$ is the magnetic moment of neighboring $j$-site magnetic atom. $C_i$ is the hyperfine coupling constants, and $n_j(i)$ is the number of the $j$-site magnetic atoms surrounding the $i$th atom.

From Eq. (1), one can see that the $^{119}$Sn hyperfine fields at the three Sn sites are governed by the spin arrangements of their surrounding Mn atoms. In all of these magnetic structures the ordering within the planes (001) is ferromagnetic, while non-collinear magnetic structures were observed between different Mn layers. Therefore, it is reasonable to assume that the value of $^{119}$Sn hyperfine field at the 2e site is the largest one. This assumption can be supported by comparing the relative absorption intensities of these three subspectra with the Ga or Ti atomic occupancy obtained by XRD results. XRD results suggest that Ti atoms prefer the 2e site for YMn$_6$Sn$_5.4$Ti$_{0.6}$, therefore the subspectrum with the smallest absorption area and the largest hyperfine field of 118 kOe is assigned to be...

Table 2

| Compounds   | Sites  | $H_{HF}$ (kOe) | $\delta$ (mm/s) $\pm 0.08$ | $\epsilon$ (mm/s) $\pm 0.08$ | $\Gamma$ (mm/s) $\pm 0.08$ | $A$ (%) $\pm 2$ |
|-------------|--------|----------------|-----------------------------|-----------------------------|-----------------------------|----------------|
| YMn$_6$Sn$_6$ | Doublet$^a$ | 0 | 1.78 | 1.16 | 1.04 | 6 |
| $\phi_1=113^\circ$ | 2c | 97 | 1.19 | 0.02 | 1.03 | 32 |
| $\phi_2=92^\circ$ | 2d | 122 | 2.42 | $-0.65$ | 1.04 | 32 |
|               | 2e | 176 | 2.03 | $-0.37$ | 1.06 | 30 |
|               | Doublet$^a$ | 0 | 2.15 | 2.19 | 1.20 | 11 |
| YMn$_6$Sn$_5.4$Ga$_{0.6}$ | 2c | 121 | 2.05 | 0.08 | 1.24 | 32 |
| $\phi_1=105^\circ$ | 2d | 179 | 2.04 | $-0.38$ | 1.01 | 20 |
| $\phi_2=52^\circ$ | 2e | 200 | 1.91 | $-0.33$ | 1.11 | 37 |
| YMn$_6$Sn$_5.4$Ti$_{0.6}$ | 2c | 56 | 1.50 | 0.96 | 1.20 | 37 |
| $\phi_1=123^\circ$ | 2d | 68 | 2.09 | $-1.70$ | 1.20 | 40 |
| $\phi_2=109^\circ$ | 2e | 118 | 2.29 | $-0.96$ | 1.20 | 23 |

For the $E_\gamma = 23.875$ keV $\gamma$ transition in $^{119}$Sn, 1 mm/s corresponds to $7.96(2) \times 10^{-8}$ eV. $H_{HF}$, $\delta$, $\epsilon$, $\Gamma$ and $A$ are the hyperfine field, center shift, quadrupole shift or splitting, linewidth (HWHM) and relative area, respectively.

$^a$ Doublet is attributed to the impurity phase.
contribution of $^{119}$Sn at 2e site. For YMn$_6$Sn$_{4.4}$Ga$_{0.6}$, the XRD refinement indicates that Ga atoms prefer to occupy 2d sites, so the subspectrum with the intermediate hyperfine field of 179 kOe can be assigned to $^{119}$Sn at 2d sites on the basis of absorption area. Finally, the subspectrum with the smallest hyperfine field is attributed to $^{119}$Sn at 2c sites.

It was known that the magnetic moments of Mn atoms in different layers are the same in magnitude, but vary in direction. By comparing the values of hyperfine fields at 2c, 2d and 2e sites, the relative angles of Mn spins in different layers can be estimated. If we assumed that the transferred hyperfine coupling constants $C_i$ all have approximately the same value, $c$, the values of hyperfine fields at 2c, 2d and 2e sites can be written from Eq. (1) as

$$H_{HF}(2c) = 3c\mu_{Mn}\sqrt{2 + 2\cos\phi_1}$$  \hspace{1cm} (2)

$$H_{HF}(2d) = 3c\mu_{Mn}\sqrt{2 + 2\cos\phi_2}$$  \hspace{1cm} (3)

$$H_{HF}(2e) = 6c\mu_{Mn}$$  \hspace{1cm} (4)

where $\phi_1$ and $\phi_2$ are the angles of Mn moments between the Mn–(R,Sn–1)–Mn and Mn–Sn3–Sn2–Sn3–Mn layers, respectively.

From Eqs. (2)–(4), the values of $\phi_1$ and $\phi_2$ can be calculated and these are listed in Table 2. These results indicate that the Mn magnetic moments change their orientation within the basal plane on going from one R sheet to another. This complex spin ordering is expected as a consequence of T-equivalent-to-Mn, Fe), J. Alloys Compd. 185 (1996) 1851–1856.

4. Summary

The effects of Ga and Ti substitutions for Sn on the Mn spin arrangements of YMn$_6$Sn$_6$ have been investigated by means of X-ray diffraction (XRD), magnetization measurement and $^{119}$Sn Mössbauer spectroscopy. XRD refinement indicates that Ga and Ti atoms demonstrate different preference of site occupancy. A few percent of Ga substitutes for Sn at the 2d sites and is found to significantly decrease the turn angle in the Mn–Sn3–Sn2–Sn3–Mn layer and thereby increases the net magnetization. This behavior is evidenced by the observed ferromagnetic behavior in YMn$_6$Sn$_{4.4}$Ga$_{0.6}$. The substitution of Ti for Sn at the 2e sites increases the turn angles of the Mn–(Y,Sn–1)–Mn and Mn–Sn3–Sn2–Sn3–Mn layers and results in ferrimagnetic behavior.

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