Vol 10 No 4, April 2001
1009-1963/2001/10(04)/0345-04

Chinese Physics

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and IOP Publishing Ltd

STRUCTURE, MAGNETIC PROPERTIES AND GIANT MAGNETORESISTANCE OF $YMn_6Sn_{6-x}Ga_x$ (x=0–0.6) COMPOUNDS*

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(Received 23 November 2000)

Structure, magnetic and transport properties of $YMn_6Sn_{6-x}Ga_x$ ($0 \leq x \leq 0.6$) compounds with a $HfFe_6Ge_6$-type structure were investigated. It was found that the Ga substitution leads to a contraction of the unit-cell volume. A transition from an antiferromagnetic to a ferromagnetic (or ferrimagnetic) state can be observed for samples ($0.1 \leq x \leq 0.2$) with increasing temperature. The antiferro–ferromagnetic transition for samples with $x \leq 0.2$ can also be induced by an external field. The required field is very low, and decreases with increasing Ga concentration. More Ga concentration ($x \geq 0.3$) leads to the samples being ferromagnetic in the whole temperature range below the Curie temperature. The Ga substitution weakens the interlayer magnetic coupling between the Mn spins. Corresponding to the metamagnetic transition, a magnetoresistance as large as 32% under a field of 5 T was observed at 5 K for the sample with x=0.2.

Keywords: structure, magnetic properties, magnetoresistance
PACC: 7525, 7530E, 7530V

I. INTRODUCTION

Rare-earth compounds of the $R\mathrm{Mn}_6$ X_6 and $R\mathrm{Mn}_2$ X_2 (R=rare earth elements and X=Sn or Ge) types have been reported to possess diversified magnetic structures and interesting magnetic properties.[1–5] All these compounds are composed of R and Mn layers alternatively stacked along the c-axis. Up to now, magnetic structures and magnetic properties of the ternary $R\mathrm{Mn}_6$ X_6 compounds have been widely investigated. It has been found that the magnetic structures and magnetic properties are very sensitive to the Mn–Mn distances, as well as the nature of the R element. Among them, the YMn_6Sn_6 compound is antiferromagnetic over the whole temperature range below the Néel temperature T_N=333 K. The magnetic structures are characterized by ferromagnetic (001) Mn planes. The interplane Mn moments through the Mn–Sn–Sn–Mn slab are always parallel, while those occurring through the Mn–(Sn–R)–Mn slab rotate with a non-constant angle.[1,6] However, the effects of the Mn–Mn interatomic distance on their magnetic structures, magnetic and electric properties are still not very clear.

A phenomenon observed in $R\mathrm{Mn}_2\mathrm{Ge}_2$ and $R\mathrm{Mn}_6\mathrm{Ge}_6$ compounds is a metamagnetic transition from an antiferromagnetic state towards a ferromagnetic state, which can be induced by an applied field.[7–9] The transition between the two different magnetic states takes place at a critical temperature (or field), and is of the first order. The mechanism leading to the metamagnetic transition was found to be extremely sensitive to the Mn–Mn distance. Corresponding to the metamagnetic transition, giant magnetoresistance (GMR) has been observed in many systems.[10–13] Though many models have been proposed to explain GMR, it is presently accepted that GMR is caused by a spin-dependent electron scattering when the magnetic structure of the sample changes.

In this paper we report the effects of Ga substitution for Sn on the structure, magnetic and transport properties of $YMn_6Sn_{6-x}Ga_x$ ($0 \leq x \leq 0.6$) series. As Y is non-magnetic, these compounds offer the possibility for a study of the Mn–Mn interactions in relation to the unit-cell volume and temperature.

II. EXPERIMENTAL DETAILS

*Project supported by the State Key Project of Fundamental Research and the National Natural Science Foundation of China (Grant No. 19804017).

$YMn_6Sn_{6-x}Ga_x$ ($0 \leq x \leq 0.6$) polycrystalline samples were synthesized by melting the constituent elements in a high-purified Ar atmosphere by the standard arc-furnace technique. The purity of the elements was better than 99.9%. An excess of Y, Mn and Ga over the stoichiometric amount was added to compensate for the mass loss during melting. All the samples were annealed at 1023 K for ten days. X-ray diffraction (XRD) studies were carried out using a Rigaku Rint 1400 diffractometer with CuK_α radiation at room temperature. The magnetization measurements for free-powder samples were carried out in the temperature range of 5–400K using a superconducting quantum interference device (SQUID) magnetometer with a maximum field of 5 T. The magnetoresistance (MR) was measured at 5K by the four-probe method in magnetic fields up to 5 T with an electric current parallel to the magnetic field.

III. RESULTS AND DISCUSSION

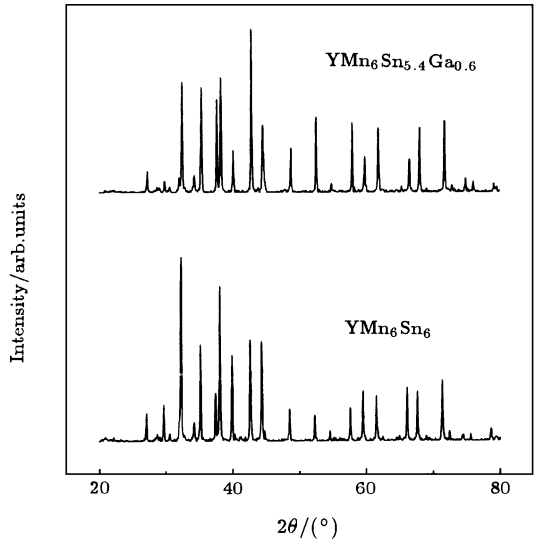


Fig.1. XRD patterns of samples with x=0 and 0.6.

All samples display peak characteristics for the $HfFe_6Ge_6$-type structure with minor Mn_3Sn_2 as an impurity phase. Figure 1 shows XRD patterns of samples with x=0 and 0.6. The lattice constants and unit-cell volumes of all compounds are shown in Fig.2. Substituting Ga for Sn atoms leads to a decrease of the lattice constants a and c due to the smaller covalent radius of Ga compared with Sn. The ratio of c/a is a constant for all samples, indicating an isotropic contraction. The Mn–Mn interatomic distance in the (001) Mn planes is equal to the lattice constant $a/2$. The Mn–Mn interatomic distance between interplanes is proportional to the lattice constant c. So the decrease of the lattice constants by substituting Ga for Sn atoms means the reduction of the Mn–Mn interatomic distances.

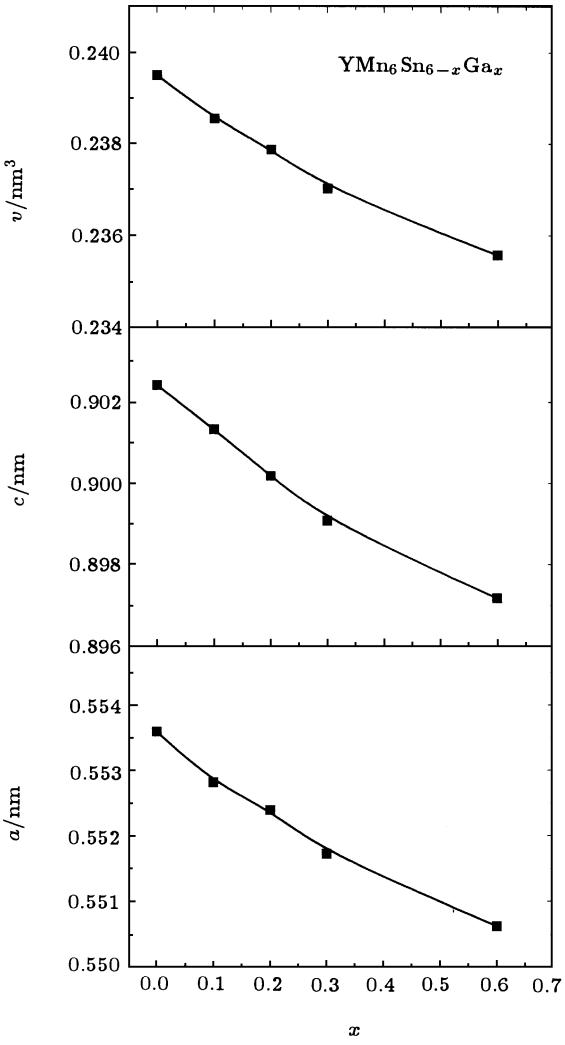


Fig.2. The lattice constants and unit-cell volumes of $YMn_6Sn_{6-x}Ga_x$ compounds.

The temperature dependence of the magnetization of $YMn_6Sn_{6-x}Ga_x$ compounds measured at a constant applied field of 0.05 T by zero field-cooling is shown in Fig.3. Three concentration regions with different magnetic properties appear: (1) for x=0, the sample has antiferromagnetic properties; (2) for $0.1 \leq x \leq 0.2$, the samples undergo a metamagnetic transition from the antiferromagnetic to the ferromagnetic state as the temperature rises; (3) for $x \geq 0.3$, the samples are ferromagnets in the whole temperature range below the Curie temperature T_c. The values of T_c increase monotonically with increasing x, reaching 367 K for x=0.6. The T_c is mainly deter-

mined by the ferromagnetic interaction in (001) Mn planes. Therefore, the small amount of Ga substitution for Sn atoms enhances the ferromagnetic interactions in Mn planes, due to the decrease of Mn–Mn interatomic distance. The small step on M–T curves near 270 K may be a contribution from the impurity Mn_3Sn_2 (Mn_3Sn_2 phase orders ferromagnetically near 270 K[1]).

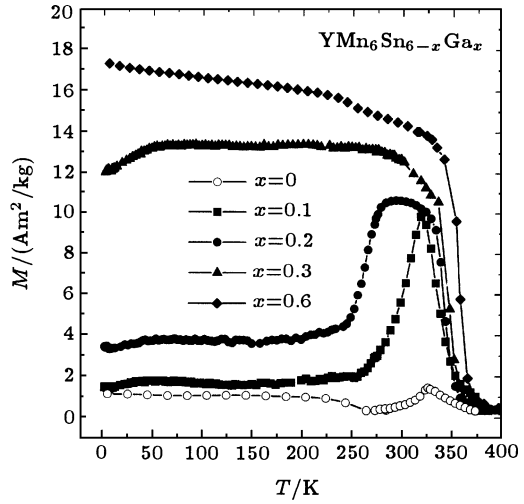


Fig.3. Temperature dependence of magnetization of $YMn_6Sn_{6-x}Ga_x$ at a magnetic field of 0.05 T.

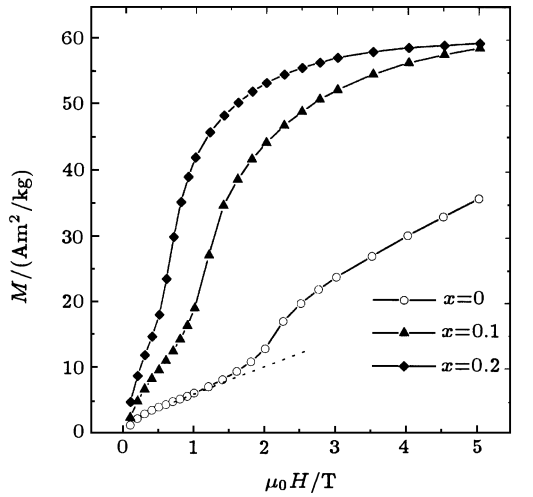


Fig.4. Magnetization of $YMn_6Sn_{6-x}Ga_x$ as a function of applied magnetic field at 5 K.

Figure 4 shows isothermal magnetization curves at 5 K for samples with $0 \leq x \leq 0.2$. It is known that YMn_6Sn_6 is a pure antiferromagnet and preliminary neutron diffraction experiments have given evidence for a helical arrangement of the Mn moments in this compound. The steep linear increase of the magnetization for x=0 below 2 T can be interpreted as a rotation of the magnetization vectors of the Mn sublattices towards the direction parallel to the applied field. It is noteworthy that the magnetization deviates upward from the linear dotted line above 2T, indicating a metamagnetic transition. The transition takes place at a relatively low magnetic field, indicating that the magnetic structure of YMn_6Sn_6 is not stable. The metamagnetic transition is also observed at 5 K for samples with x=0.1 and 0.2. The critical magnetic field of the transition decreases with increasing Ga concentration for $x \leq 0.2$. The samples with $x \geq 0.3$ exhibit ferromagnetic behaviour without any trace of metamagnetism. For simplicity, we consider three types of Mn–Mn interactions, a direct ferromagnetic interaction within the (001) Mn planes, two indirect interlayer interactions through Mn–Sn–Sn–Mn and Mn–(R,Sn)–Mn slabs, at least one of which is antiferromagnetic. Due to the helical arrangement of the Mn moments, the data presented in Fig.4 cannot be analysed in terms of the two- or three-sublattice models used for collinear magnetic structure. The effect of such a helical spin configuration on the magnetization curve is still a subject of investigation and therefore the mean-field interlayer-coupling constant cannot be derived from the slope of the linear portion on M–H curves. However, the fact that the antiferro–ferromagnetic transition is easily achieved by changing the external magnetic field or chemical composition in $YMn_6Sn_{6-x}Ga_x$ suggests the rather weak interlayer interactions. The critical field of the metamagnetic transition decreases with increasing Ga concentration, demonstrating a weakening of the interlayer antiferromagnetic interaction due to the decrease of the Mn–Mn interatomic distance, especially the Mn–Mn distance along the c-axis. The interlayer interactions through Mn–Sn–Sn–Mn slab can be altered by indirect superexchange via the conduction electrons, such as RKKY interaction. Such an effect might be related to a change within the Mn–Sn bond.

Figure 5 shows the MR of the samples with $x \leq 0.2$ at 5 K. The ratio $[\rho(H)-\rho(0)]\times 100/\rho(0)$ has been used to represent MR. The magnitude of MR for all the three samples is very small at low magnetic fields, but it increases remarkably above a critical field. The decrease of the critical magnetic field for MR with increasing Ga content corresponds well with that of the metamagnetic transition observed in the magnetization curves shown in Fig.4. An MR as large as 32% under a magnetic field of 5 T is achieved at 5 K

in the sample with x=0.2. The MR of the sample (x=0.3) without the metamagnetic transition is very small ($\leq$5%). The large MR found in $YMn_6Sn_{6-x}Ga_x$ compounds might result from the spin-flip scattering related to the metamagnetic transition. When the metamagnetic transition from an antiferromagnetic to a ferromagnetic spin configuration occurs, the Mn spins may scatter conduction electrons by means of d-p scattering.

In summary, antiferro-ferromagnetic transition from the antiferromagnet in $YMn_6Sn_{6-x}Ga_x$ compounds can be produced either by replacing part of Sn by Ga or by the application of a magnetic field. The critical magnetic field transforming the samples into the ferromagnetic state is very low, and decreases with increasing Ga concentration. Only for the samples with $0.1 \leq x \leq 0.2$, increasing temperature induces the antiferro–ferromagnetic transition. A giant MR of 32% under a magnetic field of 5 T is observed at 5 K in the sample with x=0.2, which is related to the metamagnetic transition.

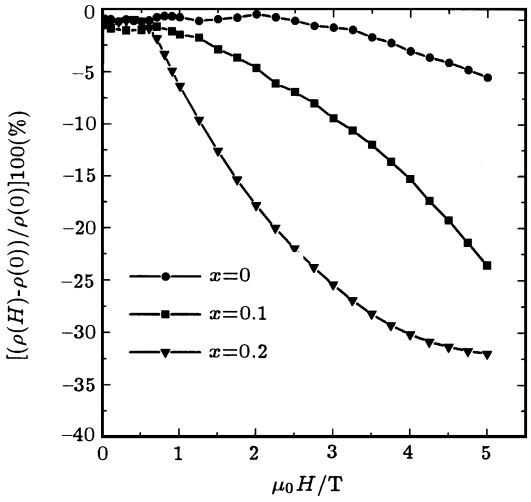


Fig.5. The magnetic field dependence of magnetoresistance of $YMn_6Sn_{6-x}Ga_x$ ($x \leq 0.2$) at 5 K.

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