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LETTER TO THE EDITOR

Magnetic anisotropy of $Sm_2Fe_{17-x}Ga_x$ compounds with $0 \leqslant x \leqslant 6$

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Abstract. The structure and intrinsic magnetic properties of $Sm_2Fe_{17-x}Ga_x$ ($0 \leqslant x \leqslant 6$) compounds were studied. These compounds crystallize in the rhombohedral Th_2Zn_{17}-type structure and are single phase except for $Sm_2Fe_{11}Ga_6$ which contains some α-Fe. The Curie temperature T_c is found first to increase and then to decrease with increasing x, attaining a maximum value of 595 K at about $x = 3$, which is 208 K higher than that of Sm_2Fe_{17}. X-ray diffraction measurements on magnetically aligned powder samples show that samples with $x < 1$ and $x > 5$ exhibit easy-plane anisotropy while samples with $1 < x < 5$ exhibit easy c-axis anisotropy. The samples with $x = 1$ and 5 show a mixture of easy-plane and easy-axis behaviour.

Since the discovery of $Sm_2Fe_{17}N_x$ [1], the search for novel hard magnetic materials has concentrated on the interstitial rare-earth iron compounds based on the 2:17-type structure. Recently, we have studied the effect of the various elemental substitutions for Fe in the $R_2Fe_{17}C_x$ alloys on their formation, structure and magnetic properties, and found that the highly stable $R_2Fe_{17}C_x$ compounds with carbon concentration up to $x = 3$ can be formed by substitution of Ga, Si etc for Fe [2–4]. In the course of study on $R_2(Fe,M)_{17}C_x$ where M = Ga, Si etc, we have also found that in the carbon-free $Sm_2Fe_{17-x}Ga_x$ compounds the substitution of Ga for Fe results in a change of room-temperature magnetocrystalline anisotropy with x, from easy plane to easy c-axis and an increase of the Curie temperature [2, 4]. In order to obtain more information about the effect of the addition of Ga on magnetic behaviour, we prepared $Sm_2Fe_{17-x}Ga_x$ compounds with $0 \leqslant x \leqslant 6$ by arc melting. In this letter, the structure and magnetic properties of $Sm_2Fe_{17-x}Ga_x$ compounds are reported.

The $Sm_2Fe_{17-x}Ga_x$ samples with $x = 0, 1, 2, 3, 4, 5$ and 6 were prepared by arc melting in an argon atmosphere of high purity. The starting materials of Sm, Fe and Ga were at least 99.9% pure. An excess of 4.5% Sm was added to compensate the mass loss due to the evaporation of Sm during melting. The arc melting ingots were wrapped by molybdenum foil, sealed in a steel tube, annealed at 1450 K for 26 h in a high-purity argon atmosphere and then quenched to room temperature. The ingots were then ground into powder. X-ray diffraction measurements on powder samples were performed using Co Kα radiation to determine the crystal structure. The aligned samples for magnetic anisotropy studies were prepared by mixing the powder with epoxy resin and then aligning in a magnetic field of 20 kOe. The Curie temperatures were determined from the temperature dependence of the magnetization measured by a vibrating sample magnetometer or a magnetic balance in a magnetic field of 1 kOe.

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Typical x-ray diffraction patterns of the $Sm_2Fe_{17-x}Ga_x$ compounds are shown in figure 1. All samples investigated are found to be single-phase compounds of the rhombohedral Th_2Zn_{17}-type structure except for $Sm_2Fe_{11}Ga_6$ which contains some α-Fe. The substitution of Ga for Fe in the Sm_2Fe_{17} compounds results in an expansion of the unit cell. Table 1 summarizes the lattice parameters a and c, and the unit cell volumes v obtained from the x-ray diffraction patterns as a function of Ga concentration. An approximately linear dependence of the unit cell volume on Ga concentration is observed. For $Sm_2Fe_{11}Ga_6$ an increase of about 6.6% in the unit cell volume is observed compared with the gallium-free compound of Sm_2Fe_{17}. This may be related to the atomic volumes of Fe and Ga. The substitution of larger Ga atoms for Fe leads to an expansion of the lattice.

Table 1. The lattice parameter a and c, unit cell volume v, Curie temperature T_c and easy magnetization direction of $Sm_2Fe_{17-x}Ga_x$.

Compound	a (Å)	c (Å)	v (Å)	T_c (K)	Anisotropy
Sm_2Fe_{17}	8.559	12.410	787.3	387	Plane
$Sm_2Fe_{16}Ga_1$	8.611	12.442	798.9	512	Cone
$Sm_2Fe_{15}Ga_2$	8.648	12.515	810.6	565	c-axis
$Sm_2Fe_{14}Ga_3$	8.673	12.577	819.3	595	c-axis
$Sm_2Fe_{13}Ga_4$	8.674	12.617	822.1	588	c-axis
$Sm_2Fe_{12}Ga_5$	8.704	12.680	831.9	525	Cone
$Sm_2Fe_{11}Ga_6$	8.733	12.709	839.4	463	Plane

It can be seen from figure 1 that the intensities of the peak in the x-ray diffraction patterns of the samples with $x \geqslant 5$ are not inconsistent with that of the samples with $x \leqslant 4$, except that the intensity of the (211) reflection for $x \geqslant 5$ shows an increase. This intensity change may result from the different occupancy site of Ga atoms, although the present results of x-ray diffraction do not provide enough information to be able to determine the occupancy site of Ga atoms. However, it has been demonstrated on the basis of the neutron diffraction studies on $Ho_2Fe_{17-x}Ga_xC_2$ [5] that the Ga atoms preferentially occupy 18h sites for $x \leqslant 4$, and 18h and 6c sites for $x > 4$.

The Ga-concentration dependence of the Curie temperature T_c of $Sm_2Fe_{17-x}Ga_x$ compounds is also listed in table 1. An important effect of the substitution of Ga for Fe in the Sm_2Fe_{17} compound on Curie temperature is observed. T_c is found first to increase and then to decrease with increasing x, having a maximum value of 595 K at about $x = 3$, as shown in figure 2. This dependence of T_c on x is very similar to that in Al-substituted R_2Fe_{17} compounds with R = Y, Ho [6] and Sm [7]. In the $Sm_2Fe_{17-x}Al_x$ compounds, the maximum value of T_c is 471 K, which is 80 K higher than that of Sm_2Fe_{17}. For $Sm_2Fe_{17-x}Ga_x$ compounds with $x = 3$, the increase of Curie temperature is about 200 K compared with the Ga-free compound with $x = 0$. This indicates that the substitution of Ga for Fe has a larger effect on the Curie temperature than the substitution of Al in the Sm_2Fe_{17} compound. In the Fe-rich rare earth iron compounds, the Curie temperature is mainly determined by the magnitude of the exchange interactions between Fe–Fe atoms, which is strongly dependent on the Fe–Fe interatomic distance. It has been evidenced in the previous studies of the magnetic properties of rare-earth iron compounds that in many cases the enhancement in Curie temperature is primarily due to the lattice expansion induced by interstitial or substitutional additions. Jacobs *et al* [6] have shown that the exchange coupling between Fe–Fe atoms in $R_2Fe_{17-x}Al_x$ increases strongly with Al substitution, and the exchange coupling between rare earth–Fe atoms is almost independent of the Al

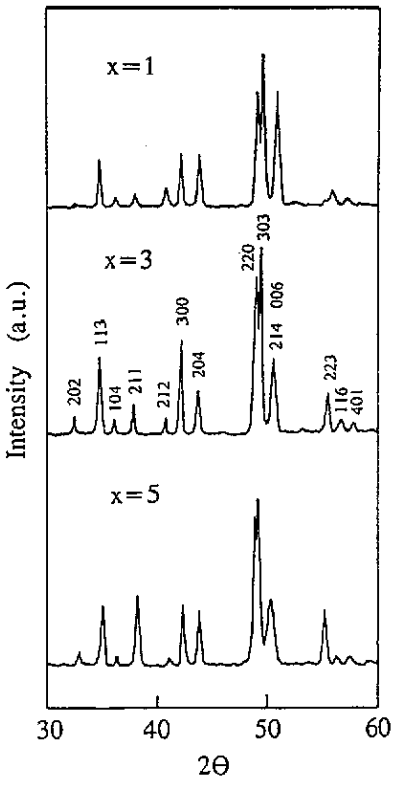

Figure 1. Co Kα x-ray diffraction patterns of $Sm_2Fe_{17-x}Ga_x$ compounds with x = 1, 3 and 5.

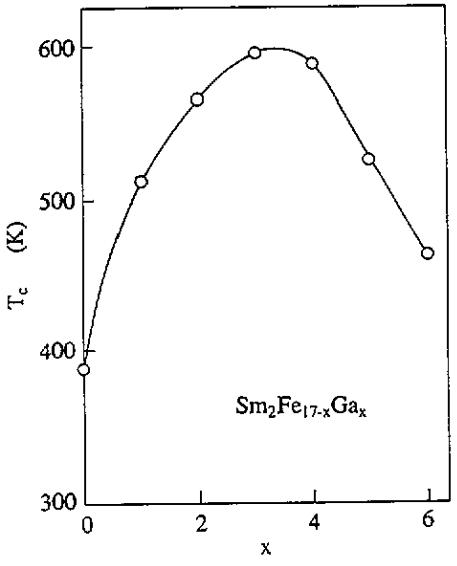

Figure 2. Ga-concentration dependence of the Curie temperature of $Sm_2Fe_{17-x}Ga_x$ compounds.

concentration. The similarity of the composition dependence of the Curie temperature in the Ga and Al substituted Sm_2Fe_{17} compounds indicates that the effects of Ga and Al on the exchange coupling are similar. The increase of T_c in $Sm_2Fe_{17-x}Ga_x$ with $x \leqslant 3$ results from the increase of Fe–Fe exchange coupling with x. The further substitution of non-magnetic Ga, when $x > 3$, would decrease the Fe–Fe exchange coupling due to the further increase of Fe–Fe distance and the decrease of the number of Fe–Fe atom pairs, resulting in the drop in T_c.

The room-temperature x-ray diffraction patterns of magnetically aligned powder samples of $Sm_2Fe_{17-x}Ga_x$ are shown in figure 3. It can be seen that the substitution of Ga for Fe in the Sm_2Fe_{17} compound has an exceptionally important effect on its magnetocrystalline anisotropy. The samples with $x < 1$ and $x > 5$ are found to have an easy-planar magnetocrystalline anisotropy. For the samples with $1 \leqslant x \leqslant 5$, a uniaxial magnetocrystalline anisotropy is observed. The samples with x = 1 and 5 show a mixture of easy-plane and easy-axis behaviour. The substitution of Ga for Fe results in a change of magnetic anisotropy from easy plane to easy c-axis, then to easy plane again. It is known that the magnetocrystalline anisotropy in the rare-earth iron compounds results from the competing anisotropy of the rare earth and iron sublattices. Usually, the rare-earth sublattice anisotropy is determined by the product of the second-order crystal field parameter A_{20} and the second-order Stevens coefficient α_J. A negative product of A_{20} and α_J gives a characteristic of uniaxial anisotropy. It has been shown previously that the second-order crystal field parameters of R_2Fe_{17} are negative, and that Sm has a positive

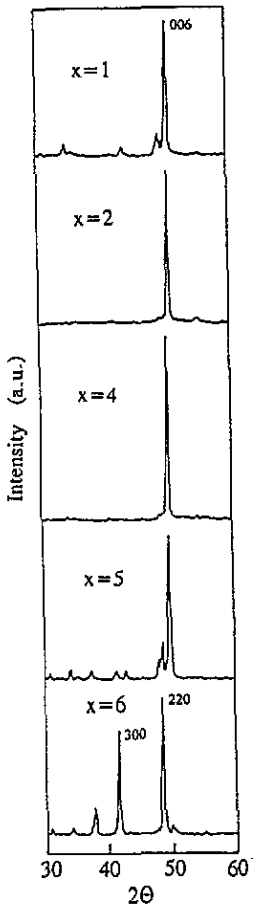

Figure 3. Co Kα x-ray diffraction patterns of magnetically aligned $Sm_2Fe_{17-x}Ga_x$ compounds with x = 1, 2, 4, 5 and 6.

second-order Stevens coefficient. The Sm sublattice in the Sm_2Fe_{17} compound has an easy c-axis anisotropy, however, the planar anisotropy of the Fe sublattice overcomes the axial anisotropy of the Sm sublattice. The present results show that the addition of an appropriate amount of Ga in the Sm_2Fe_{17} leads to a strong increase of the uniaxial anisotropy. In the Ga-substituted Sm_2Fe_{17} compounds with $2 \leqslant x \leqslant 4$, the easy c-axis anisotropy of the Sm sublattice becomes strong enough to suppress the basal plane anisotropy of the Fe sublattice at room temperature. Accordingly, it is suggested that the substitution of some Ga for Fe in the Sm_2Fe_{17} enhances the magnitude of the negative second-order crystal parameter.

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