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Magnetic properties of $Sm_2Fe_{17-x}Ga_xC_2$ compounds

Bao-gen Shen, Fang-wei Wang, Hua-yang Gong, Zhao-hua Cheng, Bing Liang, Jun-xian Zhang and Shao-ying Zhang

State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences, PO Box 603, Beijing 100080, People's Republic of China

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Abstract. The $Sm_2Fe_{17-x}Ga_xC_2$ ($x = 0, 2, 3, 4, 5$ and 6) samples were prepared by arc melting. X-ray diffraction shows that these alloys are single-phase compounds of the rhombohedral Th_2Zn_{17}-type structure except for the sample with $x = 0$, which exhibits a multiphase structure with a predominant α-Fe phase. The lattice constants a and c and the unit-cell volumes v of $Sm_2Fe_{17-x}Ga_xC_2$ compounds with $x \geqslant 2$ increase monotonically with increasing Ga concentration. The Curie temperature T_C and room-temperature saturation magnetization M_s are found to decrease monotonically with increasing Ga concentration x from 635 to 338 K and from 102.3 to 29.0 emu g^{-1} respectively, as x increases from 2 to 6. The $Sm_2Fe_{17-x}Ga_xC_2$ compounds exhibit an easy c-axis anisotropy. The room-temperature anisotropy field is 135 kOe for $x = 2$, which is about 55 kOe higher than that of $Nd_2Fe_{14}B$, and it decreases to 60 kOe at $x = 4$.

1. Introduction

The rare-earth (R) iron-rich R_2Fe_{17} intermetallic compounds are not suitable for permanent magnets because of their low Curie temperatures and room-temperature plane magnetic anisotropies. In order to increase the Curie temperatures of R_2Fe_{17} compounds and to modify their anisotropies, a number of investigations have been done. Introducing interstitial atoms into R_2Fe_{17} compounds by the gas–solid reaction [1–3] and melt-spinning [4] methods leads to a considerable enhancement of the Curie temperature. The $Sm_2Fe_{17}N_y$ and $Sm_2Fe_{17}C_y$ compounds show uniaxial anisotropy with an anisotropy of 140 kOe for nitride [5] and 150 ± 5 kOe for carbide with $y = 2.5$ [3] at room temperature. It is unfortunate that the nitrides and carbides prepared by gas–solid reaction have a poor high-temperature stability. In our previous work [6–8], it was found that the high-carbon $R_2Fe_{17}C_x$ compounds by the substitution of Ga, Al or Si exhibit a high thermal stability, in contrast with the carbides produced by the gas–solid reaction. We have studied [6–9] the formation, structure and magnetic properties of the arc-melted compounds of $R_2(Fe, M)_{17}C_x$ (R ≡ Y, Nd, Sm, Gd, Tb, Dy, Ho, Er or Tm; M ≡ Ga, Al or Si; $x \leqslant 3.0$) with the rhombohedral Th_2Zn_{17}-type or hexagonal Th_2Ni_{17}-type structures and found that the arc-melted $Sm_2(Fe, M)_{17}C_x$ compounds with $x \geqslant 1.5$ and relatively lower M concentration have a Curie temperature of higher than 600 K and a room-temperature anisotropy field of higher than 90 kOe. A high coercivity of 15 kOe at room temperature was obtained for the $Sm_2(Fe, Ga)_{17}C_x$ compounds by melt spinning [6, 10]. The important effect on the magnetic properties of R_2Fe_{17} compounds results not only from the addition of interstitial carbon atoms but also from the substitution of Ga, Al, Si, Co, Ni, etc, for Fe [11–16]. The substitution of Ga in Sm_2Fe_{17} can cause a room-temperature uniaxial anisotropy to develop without

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introducing interstitial carbon or nitrogen atoms [15, 16]. In our previous papers, the effect of the interstitial carbon atoms on the structure and magnetic properties of $Sm_2(Fe, Ga)_{17}C_x$ compounds was presented [6, 7]. Here, we report the influence of the substitution of Ga for Fe on the structure and magnetic properties of $Sm_2Fe_{17}C_2$.

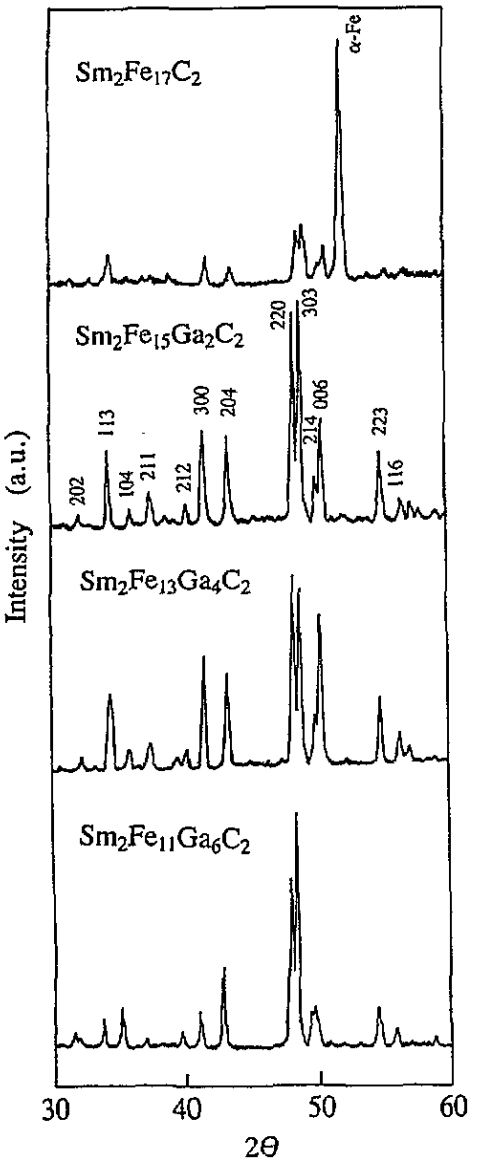

Figure 1. X-ray diffraction patterns of arc-melted $Sm_2Fe_{17-x}Ga_xC_2$ annealed at 1450 K for 48 h with $x = 0$, for 96 h with $x = 2$, and for 12 h with $x = 4$ and 6 (a.u., arbitrary units).

2. Experimental details

The $Sm_2Fe_{1-x}Ga_xC_2$ alloys with $x = 0$, 2, 3, 4, 5 and 6 were prepared by arc melting in an argon atmosphere of high purity. The raw materials of Sm, Fe, Ga and Fe–C alloy were at least 99.9% pure. The ingots were melted at least four times to ensure homogeneity. An

Table 1. Structure and magnetic parameters of $Sm_2Fe_{17-x}Ga_xC_2$ compounds.

Compound	a (Å)	c (Å)	v (Å^3)	T_C (K)	M_s (1.5 K) (emu g^{-1})	M_s (300 K) (emu g^{-1})	H_A (1.5 K) (kOe)	H_A (300 K) (kOe)
$Sm_2Fe_{15}Ga_2C_2$	8.724	12.586	829.4	635	114.2	102.3	> 250	135
$Sm_2Fe_{14}Ga_4C_2$	8.747	12.621	836.2	629	99.2	88.7	> 250	93
$Sm_2Fe_{13}Ga_4C_2$	8.754	12.654	839.8	539	86.7	73.1	> 250	60
$Sm_2Fe_{12}Ga_5C_2$	8.778	12.674	845.7	432	71.0	52.9		
$Sm_2Fe_{11}Ga_6C_2$	8.798	12.721	852.7	338	57.9	29.0		

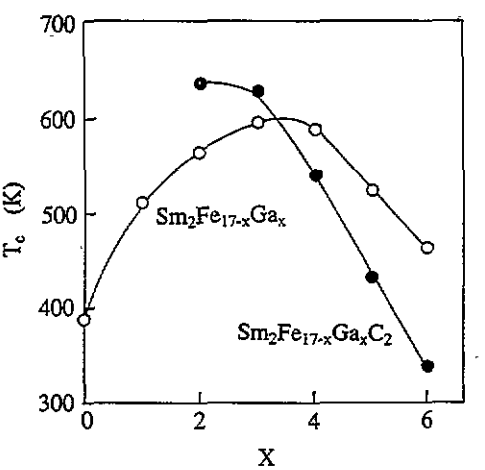

Figure 2. Curie temperature T_C for $Sm_2Fe_{17-x}Ga_x$ [6] and $Sm_2Fe_{17-x}Ga_xC_2$ as a function of Ga concentration.

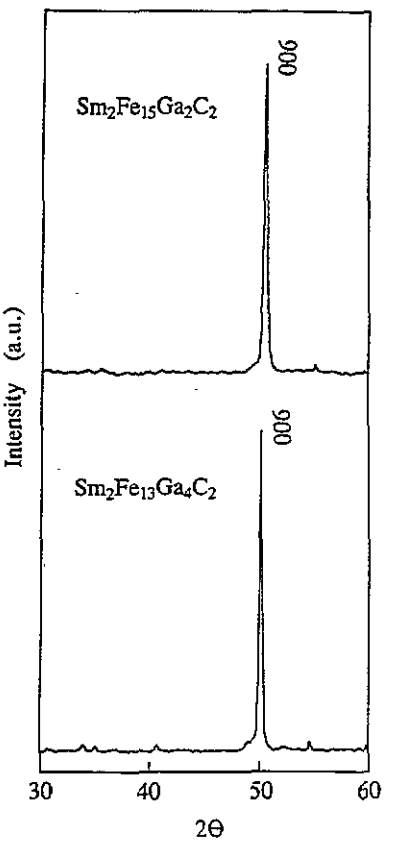

Figure 3. X-ray diffraction patterns of magnetically aligned $Sm_2Fe_{17-x}Ga_xC_2$ powder samples with $x = 2$ and 4 (a.u., arbitrary units).

excess of 4.5% Sm was added to compensate for the evaporation of Sm during melting. The heat treatment of the arc-melted ingots was performed in a steel tube in a highly purified argon atmosphere at 1450 K for 12–96 h. X-ray diffraction measurements on powder samples were performed using Co Kα radiation to identify the single phase and to determine the crystallographic structure. The room-temperature saturation magnetization was measured with an extracting-sample magnetometer in a field of 65 kOe. The Curie temperatures were determined from the temperature dependence of magnetization measured with a vibrating-sample magnetometer and a magnetic balance in a magnetic field of 1 kOe. The aligned samples for anisotropy field measurements were prepared by mixing the powder

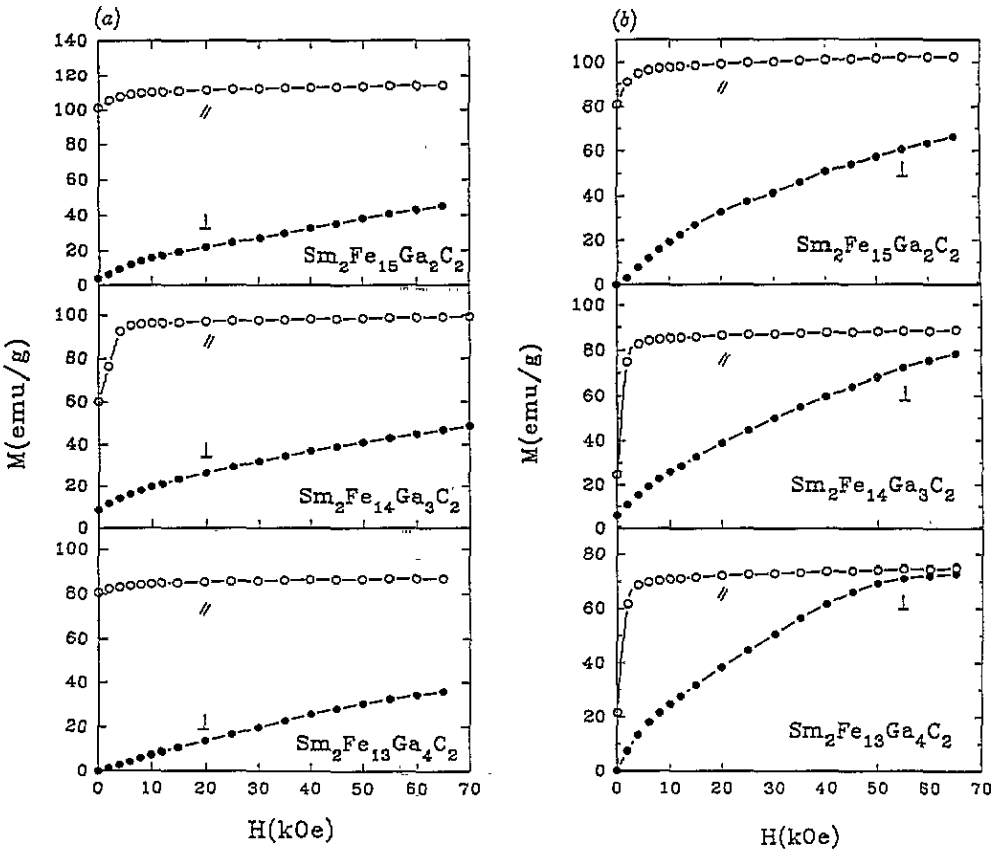

Figure 4. The magnetization curves of the oriented $Sm_2Fe_{17-x}Ga_xC_2$ ($x = 2$, 3 and 4) samples measured along and perpendicular to the aligned directions at (a) 1.5 K and (b) room temperature.

with epoxy resin and then aligning in a magnetic field of 10 kOe. The anisotropy field was determined from magnetization curves measured along and perpendicular to the orientation direction by using the extracting-sample magnetometer with a magnetic field of up to 65 kOe at 1.5 K and room temperature.

3. Results and discussion

The x-ray diffraction patterns of $Sm_2Fe_{17-x}Ga_xC_2$ with $x = 0, 2, 4$ and 6 are shown in figure 1. It is difficult to obtain a single-phase $Sm_2Fe_{17-x}Ga_xC_2$ compound with $x < 2$ by arc melting. X-ray diffraction, as shown in figure 1, of $Sm_2Fe_{17}C_2$ shows a multiphase structure with a predominant α-Fe phase. For $x = 2$, the sample consists of the 2:17 phase and α-Fe phase; however, high-temperature annealing leads to the formation of the 2:17-type single phase. Our study shows that the arc-melted $Sm_2Fe_{17-x}Ga_xC_2$ alloys with $x \geqslant 3$ are single phase with the rhombohedral Th_2Zn_{17}-type structure. No significant diffraction from the impurity phase is observed. The partial substitution of Ga for Fe helps the formation of the high-carbon rare-earth iron compounds with 2:17-type structure, as shown in our previous work [6, 7, 9].

Table 1 summarizes the lattice constants a and c and the unit-cell volumes v obtained from the x-ray diffraction patterns of $Sm_2Fe_{17-x}Ga_xC_2$ compounds with $2 \leqslant x \leqslant 6$ as a function of Ga concentration. The substitution of Ga for Fe was found to increase the unit-cell volumes. An approximately linear dependence of the unit-cell volume on Ga concentration is observed. For $Sm_2Fe_{17-x}Ga_xC_2$, the unit-cell volume expansion is about 2% compared with those of $Sm_2Fe_{17-x}Ga_x$ compounds with the same Ga concentration [15]. The increase in cell volume per Ga atom is found to be about 5.4 Å^3.

The saturation magnetization M_s and Curie temperature T_C for $Sm_2Fe_{17-x}Ga_xC_2$ compounds are also listed in table 1. The saturation magnetization at 1.5 K was found to decrease with increasing x from 114.2 emu g^{-1} for $x = 2$ to 57.9 emu g^{-1} for $x = 6$. The room-temperature saturation magnetization is in excess of 100 emu g^{-1} for $x = 2$ and it decreases monotonically with increasing Ga content to 29.0 emu g^{-1} at $x = 6$. T_C for $Sm_2Fe_{17-x}Ga_xC_2$ with $x \leqslant 3$ is found to be about 630 K, which is about 240 K higher than that of Sm_2Fe_{17} and about 130 K higher than that of $Sm_2Fe_{17}C$ [12]. However, a higher Ga concentration ($x > 3$) decreases T_C. Figure 2 shows the Curie temperatures T_C of $Sm_2Fe_{17-x}Ga_x$ [15] and $Sm_2Fe_{17-x}Ga_xC_2$ compounds as a function of Ga concentration, for comparison. It is shown that, when the Ga concentration x is lower than 3, the introduction of interstitial carbon atoms or the substitutional Ga atoms in Sm_2Fe_{17} leads to a strong increase in Curie temperature. The enhancement of T_C is mainly due to the lattice expansion. However, a decrease in Curie temperature with increasing x in the Ga-substituted compounds with $x > 3$ is observed, although the further substitution of Ga results in a monotonic increase in the unit-cell volume. In general, in the Fe-rich rare-earth–iron compounds the Curie temperature is mainly determined by the Fe–Fe exchange interactions, which is strongly dependent on the interatomic distance. The larger substitution of larger Ga for Fe, when $x > 3$, would decrease the Fe–Fe exchange interactions owing to the further increase in Fe–Fe distance and the decrease in the number of Fe–Fe atom pairs, resulting in a drop in T_C.

The $Sm_2Fe_{17-x}Ga_xC_2$ samples with $x \leqslant 6$ are found to have a strong easy c-axis anisotropy at room temperature. This can be clearly seen from the x-ray diffraction patterns of magnetically aligned powder samples, as is shown in figure 3, for $Sm_2Fe_{17-x}Ga_xC_2$ with $x = 2$ and 4. The strong enhancement in the (0, 0, 6) reflection and the absence of $(h, k, 0)$ indicates that the c axis is the easy direction of magnetization. The strong uniaxial magnetocrystalline anisotropy of the Sm sublattice in the $Sm_2Fe_{17-x}Ga_xC_2$ can result from the effect of both Ga and C additions. It has been shown previously that the substitution of Ga for Fe in Sm_2Fe_{17} leads to a change in magnetocrystalline anisotropy from the basal plane to the c axis at room temperature [15, 16]. The addition of carbon atoms causes a large negative crystal-field parameter A_{20} [17] and, in consequence, increases the anisotropy of the Sm sublattice. Figure 4 shows the magnetization curves of $Sm_2Fe_{17-x}Ga_xC_2$ ($x = 2$, 3 and 4) compounds measured along and perpendicular to the aligned directions at 1.5 K and room temperature. The magnetocrystalline anisotropy field H_A estimated from magnetization curves is listed in table 1. H_A is found to be in excess of 250 kOe at 1.5 K. The room-temperature anisotropy field of the sample with $x = 2$ is 135 kOe, which is 55 kOe higher than that of $Nd_2Fe_{14}B$. The further substitution of non-magnetic Ga ($x > 2$) decreases H_A.

Acknowledgments

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References

[1] Coey J M D and Sun H 1990 *J. Magn. Magn. Mater.* **87** L251
[2] Coey J M D, Sun H, Otani Y and Hurley D P F 1991 *J. Magn. Magn. Mater.* **98** 76
[3] Liao L X, Chen X, Altounian Z and Ryan D H 1992 *Appl. Phys. Lett.* **60** 129
[4] Cao L, Kong L S and Shen B G 1992 *J. Phys.: Condens. Matter* **4** L515
[5] Katter M, Wecker J, Schultz L and Grossinger R 1990 *J. Magn. Magn. Mater.* **92** L14
[6] Shen B G, Kong L S, Wang F W and Cao L 1993 *Appl. Phys. Lett.* **63** 2288
[7] Shen B G, Wang F W, Kong L S, Cao L and Zhan W S 1994 *J. Appl. Phys.* **75** 6253
[8] Cheng Z H, Shen B G, Wang F W, Zhang J X, Gong H Y and Zhao J G 1994 *J. Phys.: Condens. Matter* **6** L185
[9] Shen B G, Wang F W, Kong L S, Cao L and Guo H Q 1993 *J. Magn. Magn. Mater.* **127** 1267
[10] Kong L S, Shen B G, Wang F W, Cao L, Guo H Q and Ning T S 1994 *J. Appl. Phys.* **75** 6250
[11] Alp E E, Umarji A M, Malik S K, Snenoy G K, Huang M Q, Boltich E B and Wallace W E 1987 *J. Magn. Magn. Mater.* **68** 305
[12] Weitzer F, Hiebl K and Rogl P 1990 *IEEE Trans. Magn.* **MAG-26** 2661
[13] Hu B P, Rao X L, Xu J M, Liu G C, Dong X L, Li H, Yin L and Zhao Z R 1992 *J. Magn. Magn. Mater.* **114** 138
[14] Jacobs T H, Buschow K H J, Zhou G F, Li X and de Boer F R 1992 *J. Magn. Magn. Mater.* **116** 220
[15] Shen B G, Wang F W, Kong L S and Cao L 1993 *J. Phys.: Condens. Matter* **5** L685
[16] Wang Z and Dunlap R A 1994 *Phil. Mag.* B **69** 103
[17] Jacobs T H, Dirken M W, Thiel R C, de Jongh L J and Buschow K H J 1990 *J. Magn. Magn. Mater.* **83** 293