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The change in magnetic anisotropy in $R_2Fe_{17-x}Al_x$ compounds (R = Sm or Tb)

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Abstract. The structure and magnetic properties of $R_2Fe_{17-x}Al_x$ compounds with R = Sm and with R = Tb were studied. The compounds have a rhombohedral Th_2Zn_{17}-type structure. The Curie temperature T_C is found first to increase and then to decrease with increasing x. The substitution of Al has a significant effect on the magnetic anisotropy of $R_2Fe_{17-x}Al_x$ compounds. A small amount of Al substitution in Sm_2Fe_{17} induces uniaxial anisotropy and a further substitution results in a change in the easy-magnetization direction from c axis to basal plane, whereas an opposite change in the magnetocrystalline anisotropy has been observed in the alloys of $Tb_2Fe_{17-x}Al_x$. The samples of $Tb_2Fe_{17-x}Al_x$ with $x \geqslant 6$ exhibit an easy c axis anisotropy.

The values of Curie temperature for R_2Fe_{17} are relatively low and none of them exhibits an easy c axis anisotropy at room temperature. These drawbacks restrict the possible application of these materials as permanent magnets. In order to improve their intrinsic magnetic properties, much work has been done by either substituting other elements in the R_2Fe_{17} structure [1–4] or preparing materials with interstitial atoms [5–7] or both. Recently, it was discovered that the substitution of Ga, Al, Si, etc, for Fe not only could facilitate the formation of $R_2Fe_{17}C_x$ compounds with high-carbon concentration but also could increase the anisotropy field of $Sm_2Fe_{17}C_x$ [8–10]. In previous work [11], we found that substitution of a small amount of Al resulted in a rapid increase in the anisotropy field, but further substitution decreased the anisotropy field. In order to obtain more direct information concerning the effect of Al substitution on magnetic anisotropy, we prepared the parent alloys $Sm_2Fe_{17-x}Al_x$ and $Tb_2Fe_{17-x}Al_x$ with $0 \leqslant x \leqslant 7$ by arc melting. In this paper, the structure and magnetic anisotropy of $R_2Fe_{17-x}Al_x$ (R = Sm or Tb) are reported.

The samples of $Sm_2Fe_{17-x}Al_x$ and $Tb_2Fe_{17-x}Al_x$ with x = 0, 2, 4, 5, 6 and 7 were arc melted in a high-purity argon atmosphere. The ingot alloys were remelted at least four times to ensure homogeneity and then annealed under an argon atmosphere at 1273 K for 24 h, followed by quenching into water. X-ray diffraction experiments were performed on powder samples using Cu Kα radiation to determine the phase structure and the lattice parameters. The values of Curie temperature T_C and compensation temperature T_{comp} were derived from the temperature dependence of the magnetization $M(T)$ curves measured with a vibrating-sample magnetometer in a magnetic field of 1000 Oe.

X-ray diffraction patterns demonstrate that all investigated samples crystallize in a rhombohedral Th_2Zn_{17}-type structure. The lattice constants a and c and the unit-cell volumes v for $Sm_2Fe_{17-x}Al_x$ and $Tb_2Fe_{17-x}Al_x$ are listed in table 1 and table 2, respectively. It can

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Table 1. The structural and magnetic parameters for $Sm_2Fe_{17-x}Al_x$ compounds.

x	a (Å)	c (Å)	V (Å^3)	c/a	Easy-magnetization direction
0	8.540	12.43	785	1.455	Plane
2	8.613	12.478	809.8	1.450	c axis
4	8.654	12.581	816.0	1.453	c axis
5	8.718	12.682	834.8	1.453	c axis
6	8.742	12.706	841.0	1.453	Cone
7	8.782	12.756	851.97	1.453	Plane

Table 2. The Structural and magnetic parameters for $Tb_2Fe_{17-x}Al_x$ compounds.

x	a (Å)	c (Å)	V (Å^3)	c/a	μ_{Tb} (μ_B)	T_{comp} (K)	Easy-magnetization direction
0	8.521	12.412	780.4	1.457	9.9		Plane
2	8.561	12.525	794.9	1.463	9.1		Plane
4	8.630	12.576	811.3	1.457	8,7		Plane
5	8.655	12.610	818.3	1.457	8.1		Plane
6	8.697	12.646	828.3	1.455	7.9		c axis
7	8.703	12.654	830.1	1.454	7.3	54.8	c axis

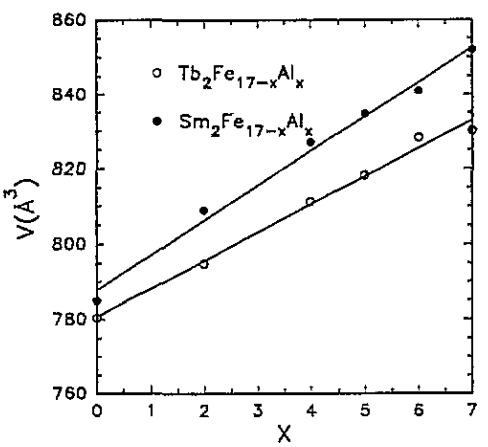

Figure 1. The unit-cell volumes v of $Sm_2Fe_{17-x}Al_x$ and $Tb_2Fe_{17-x}Al_x$ compounds as functions of Al concentration x.

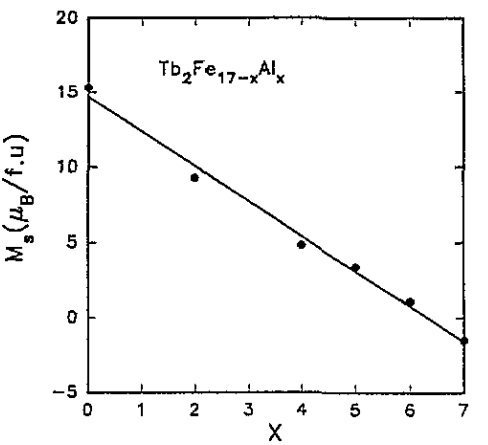

Figure 2. The Al concentration dependence of saturation magnetization M_s for $Tb_2Fe_{17-x}Al_x$ compounds.

be seen that these values show a linear dependence on Al concentration as we would expect on the basis of a simple model involving atomic volumes. The substitution of larger Al atoms for smaller Fe atoms results in the lattice expansion at a rate of 7 Å^3 per Al atom, as can be seen in figure 1. The fact that the ratio c/a is, within experimental uncertainty, independent of the Al concentration indicates that the lattice expansion is essentially an isotropic process.

The magnetization curves were measured at 1.5 K using an extracting-sample magnetometer in a magnetic field range from 0 to 70 kOe. The saturation magnetizations were obtained by fitting the experimental data of $M(H)$ versus H using the law of approach to saturation. The saturation magnetizations of $Tb_2Fe_{17-x}Al_x$ compounds as a function of Al concentration are shown in figure 2. It is found that the saturation magnetization decreases linearly with increasing Al concentration. The decrease in saturation magnetization is faster than in the case of simple magnetic dilution. The magnetic moments μ_s of $Tb_2Fe_{17-x}Al_x$

can be described as

$$\mu_s = (17 - x)\mu_{Fe} - 2\mu_{Tb}. \tag{1}$$

If we use the values of average iron magnetic moment of $Y_2Fe_{17-x}Al_x$ [12] to replace those of $Tb_2Fe_{17-x}Al_x$ compounds corresponding to the same Al concentration, the magnetic moments μ_{Tb} of Tb can be obtained, which are also summarized in table 2. The Tb magnetic moments are found to be slightly lower than the free-ion magnetic moment in the samples with a higher Al concentration. This may be due to the crystal effect which reduces μ_R or which brings about a saturation where μ_R and μ_{Fe} are non-collinear.

With increasing Al concentration, the T-sublattice moment will decrease. For the sample with $x \simeq 6.5$, it is about equal to the Tb sublattice moment at 1.5 K. At higher Al concentrations, the T sublattice moment is lower than the Tb sublattice at 1.5 K. However, because the Tb sublattice moment decreases more rapidly as function of temperature than the T sublattice moment does, they will be equal to a certain temperature.

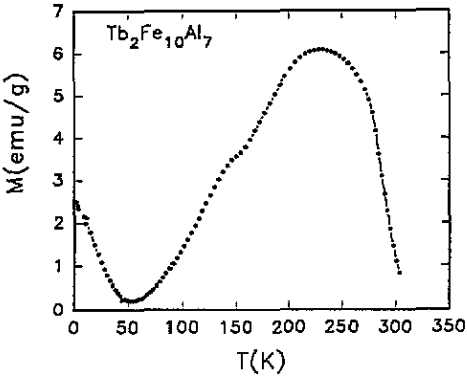

Figure 3. The temperature dependence of magnetization of $Tb_2Fe_{10}Al_7$ measured at a magnetic field of 1000 Oe.

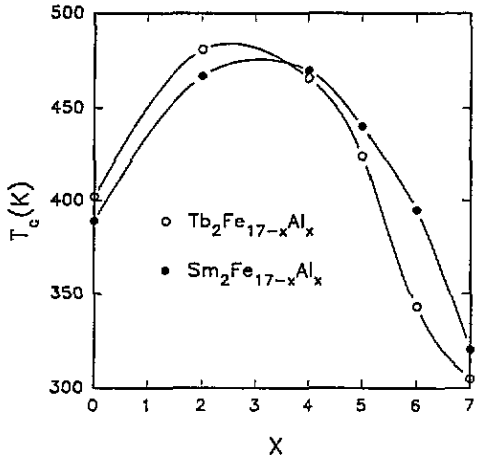

Figure 4. The Curie temperature for $Sm_2Fe_{17-x}Al_x$ and $Tb_2Fe_{17-x}Al_x$ compounds versus Al concentration x.

In order to measure the values of Curie temperature and compensation temperature more precisely, the temperature dependence of the magnetization of $Tb_2Fe_{10}Al_7$ was measured in a low magnetic field of 1000 Oe, as shown in figure 3. In general, it proved necessary to cool the samples from room temperature to 1.5 K in a high magnetic field, since otherwise the net magnetization at low temperatures vanishes or is very low. The origin of this magnetohistory effect lies in the strongly temperature-dependent anisotropy. It can be seen that the temperature at which the two sublattice moments cancel is higher for higher Al concentrations. The compensation temperature is also listed in table 2.

Figure 4 illustrates the Curie temperatures of $Sm_2Fe_{17-x}Al_x$ and $Tb_2Fe_{17-x}Al_x$ compounds as a function of Al concentration. The Al concentration dependence of Curie temperature T_C is very similar to those of $Sm_2Fe_{17-x}Ga_x$ [13] and $R_2Fe_{17-x}Al_x$ with R = Y or Ho [12]. The Curie temperatures of $Sm_2Fe_{15}Al_2$ and $Tb_2Fe_{15}Al_2$ are about 80 K higher than those of Sm_2Fe_{17} and Tb_2Fe_{17}, while for $Sm_2Fe_{17-x}Ga_x$ the increase in T_C is about 200 K compared with the Ga-free compound with $x = 0$. The Curie temperature for Fe-rich rare-earth–Fe compounds is mainly determined by the Fe–Fe exchange interaction. It

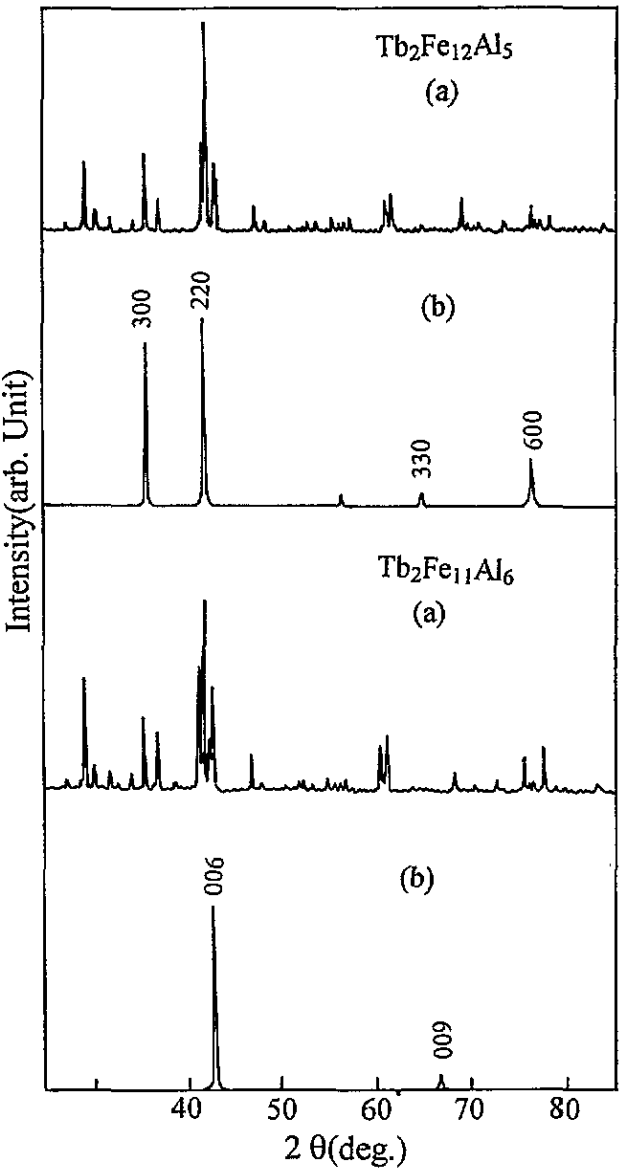

Figure 5. Room-temperature Cu Kα radiation x-ray diffraction patterns of (a) unoriented and (b) oriented samples of $Tb_2Fe_{17-x}Al_x$ with $x = 5$ and 6.

is strongly dependent on the Fe–Fe interatomic distance. The increase in Curie temperature corresponds to an increase in the positive Fe–Fe exchange coupling as a result of increasing interatomic distance. The smaller effect of Al substitution on the Curie temperature compared with that of Ga substitution may be due to substitution of the smaller Al atoms and, as a consequence, a smaller Fe–Fe distance in $R_2Fe_{17-x}Al_x$ compounds.

In order to investigate the effect of Al substitution on the magnetocrystalline anisotropy of $Sm_2F_{17-x}Al_x$ and $Tb_2Fe_{17-x}Al_x$ compounds, the samples were ground to fine powders, mixed with epoxy resin and then oriented in an applied field of 10 kOe. It is well known that x-ray diffraction studies of magnetically aligned powders can provide information about the

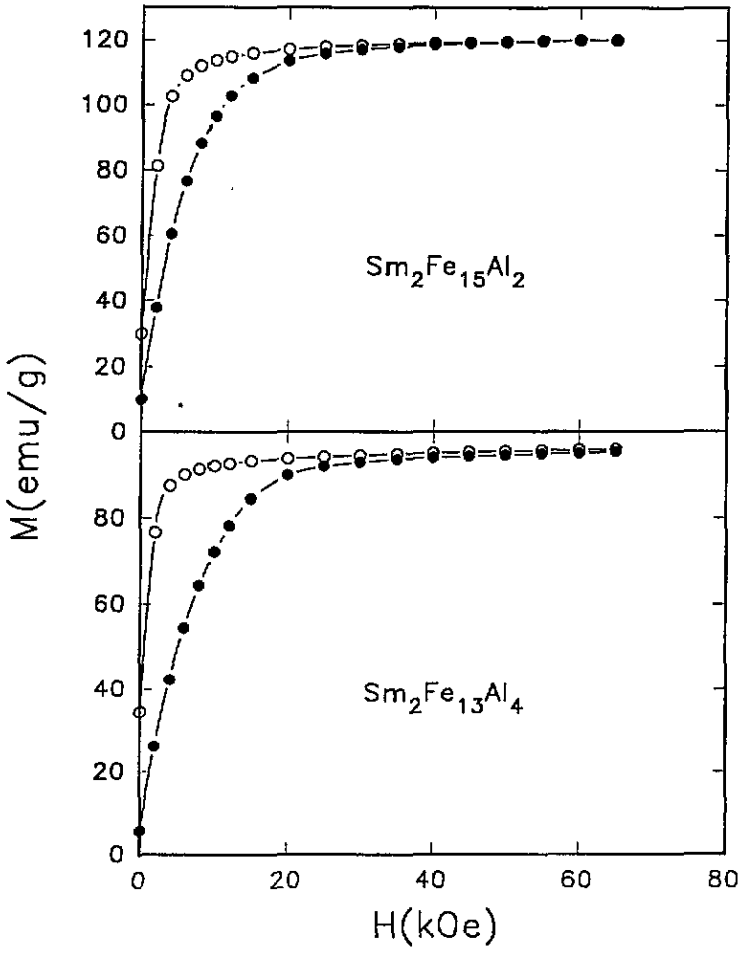

Figure 6. The magentization curves of $Sm_2Fe_{15}Al_2$ and $Sm_2Fe_{13}Al_4$ powders along (○) and perpendicular (●) to the aligned direction.

magnetocrystalline anisotropy on the basis of the diffraction peaks present in each of pattern. Figure 5 gives the room-temperature x-ray diffraction patterns of unoriented and oriented $Tb_2Fe_{17-x}Al_x$ powder samples with $x = 5$ and 6. It can be seen that the substitution of Al in R_2Fe_{17} has a significant effect on the magnetocrystalline anisotropy. Uniaxial magnetocrystalline anisotropy has been induced by the addition of larger amounts of Al ($x \geqslant 6$) into Tb_2Fe_{17}. From figure 5, it can be clearly seen that the rhombohedral c axis of $Tb_2Fe_{11}Al_6$ is readily aligned in the direction of applied field.

In contrast with $Tb_2Fe_{17-x}Al_x$ compounds, the anisotropy change of $Sm_2Fe_{17-x}Al_x$ compounds shows the opposite behaviour. The addition of a small amount of Al ($2 \leqslant x \leqslant 5$) causes uniaxial anisotropy to develop. For example, figure 6 presents the magnetization curves of $Sm_2Fe_{15}Al_2$ and $Sm_2Fe_{13}Al_4$ powders along and perpendicular to the aligned direction using an extracting-sample magnetometer with a magnetic field up to 70 kOe. The values of anisotropy fields are estimated to be about 16 kOe and 18 kOe, respectively. The sample with $x = 6$ shows a mixture of easy-plane and easy-axis behaviour. The further substitution of Al for Fe leads to a change in magnetic anisotropy from easy axis to easy

plane again. Although Wang and Dunlap [14] have reported that the substitution of a smaller amount of Al resulted in uniaxial anisotropy [14], which is similar to our results, this paper gives the first report of the anisotropy change from easy c axis to easy plane again with increasing Al concentration.

It is well known that the net anisotropy in rare-earth–Fe intermetallics is determined by the sum of the Fe sublattice and the rare-earth sublattice anisotropies. In R_2Fe_{17} compounds, the magnetization of the Fe sublattice exhibits planar anisotropy. The rare-earth sublattice anisotropy can be described by the produce of the second-order Stevens coefficient α_J and the second-order crystal parameter A_{20} on the basis of the single-ion model [15]. A negative produce $\alpha_J A_{20}$ gives a uniaxial contribution from the rare-earth sublattice to the total anisotropy. In the case of R_2Fe_{17} compounds, $A_{20} < 0$, with $\alpha_J > 0$ for R = Sm and $\alpha_J < 0$ for R = Tb. The substitution of a smaller amount of Al in Sm_2Fe_{17} increases the uniaxial anisotropy, suggesting an increase in the magnitude of the negative A_{20} and, consequently, an increase in the anisotropy of the Sm sublattice. However, when $x \geqslant 6$, $Tb_2Fe_{17-x}Al_x$ samples exhibit an easy c axis anisotropy and the anisotropy of $Sm_2Fe_{17-x}Al_x$ changes from easy c axis to easy plane. A similar change has also been observed for $R_2Fe_{17-x}Ga_x$ with R = Sm or Tb [13, 16]. Both Ga and Al have similar effects on the magnetic anisotropy of Tb_2Fe_{17} compounds. The results imply a reversal in the sign of the second-order crystal parameter A_{20} from negative to positive with increasing Al concentration. A further study of this mechanism is in progress.

Acknowledgments

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