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Magnetic properties of $SmTbFe_{17-x}Ga_x$ $[0 \leqslant x \leqslant 8]$

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Magnetic properties of $(Sm_{0.5}Tb_{0.5})_2Fe_{17-x}Ga_x$ [mentioned subsequently as $SmTbFe_{17-x}Ga_x$] have been investigated. All the compounds have formed in rhombohedral Th_2Zn_{17} structure with traces of α-Fe. The saturation magnetization decreases with increasing x and is attributed to the modification of the density of states of $3d$ band due to the low lying $3p$ band of Ga. The increase of Curie temperature up to $x=3$ and subsequent decrease is explained on the basis of band narrowing. The easy direction of magnetization is in the ab plane up to $x=5$ and changes along the c axis for $x=7$. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1855705]

I. INTRODUCTION

Rare-earth iron compounds based on the hexagonal and rhombohedral 2:17-type structure have low Curie temperatures and planar anisotropy at room temperature both of which and magnetization have been reported to improve upon the interstitial modification by nitrogen or carbon.[1,2] The thermal stability and Curie temperatures have been improved substantially by the substitution of nonmagnetic atoms such as Al, Ga, Si, for Fe.[3–8] Interstitial modification with nitrogen[9] and carbon[8] has been reported to increase magnetization and Curie temperature. Ingersoll *et al.*[10] have reported that substitution of Ga and Si for Fe in $ErPrFe_{17}$ has increased T_C until Ga/Si=3.5. Rama Rao *et al.*[11] have reviewed the magnetic properties vis-à-vis preferential occupancy of magnetic and nonmagnetic substituent elements for Fe in 2:17 compounds. In addition to modifying the Fe sublattice anisotropy, the total anisotropy may also be modified using two rare-earth elements. The purpose of this paper is to present the effect of Ga substitution on the structural and magnetic properties of $SmTbFe_{17}$.

II. EXPERIMENTAL DETAILS

All the compounds were prepared by arc melting the stochiometric amounts of starting elements (Sm and Tb, 99.9% pure; Fe, 99.95% pure; Ga, 99.99% pure) in argon atmosphere. The ingots were melted several times to ensure the homogeneity. The loss in the rare-earth element upon melting has been compensated for with 15% of Sm and 3% of Tb in excess so that the net weight loss after melting is less than 0.5%. The ingots were wrapped by tantalum foil, sealed in quartz tube at vacuum (pressure $< 10^{-5}$torr) and annealed at 950 °C in high pure argon atmosphere for seven days and quenched in ice water mixture. X-ray diffraction (XRD) measurements on powder samples were performed using Fe $K\alpha$ radiation. Magnetization measurements were carried out using a vibrating sample magnetometer (PAR make, Model no. 155). The samples for the anisotropy studies have been prepared as discussed in Ref. 10.

III. RESULTS AND DISCUSSION

The powder XRD patterns of the $SmTbFe_{17-x}Ga_x$ compounds are shown in Fig. 1(a). Si is used as a standard in order to take care of the instrumental errors in the angles. All the compounds have been formed in the rhombohedral Th_2Zn_{17}-type structure with traces of α-Fe, with the amount becoming not detectable at higher concentrations of Ga. Both the lattice parameters a and c and therefore the volume increase with Ga concentration (Table I) in accordance with the relative atomic radii of Ga and Fe.[3–11]

The magnetizations in all the compounds have reached almost their respective saturation values (M_s) which were obtained from the Honda plots [M vs $1/H$ plots near saturation where M is expanded in powers of $1/H$ and higher order terms are neglected near saturation; M_s corresponds to $1/H=0$]. The value of M_s decreases with increase in Ga concentration (Fig. 2). The magnetization in a R_2Fe_{17} compound has strong dependence on the volume similar to the magnetovolume effect in fcc iron.[9] Interstitial modification by N or C has been reported to expand the lattice and subsequently cause an increase in the magnetization and Curie temperature.[1,2] However, in the present case, while the lattice

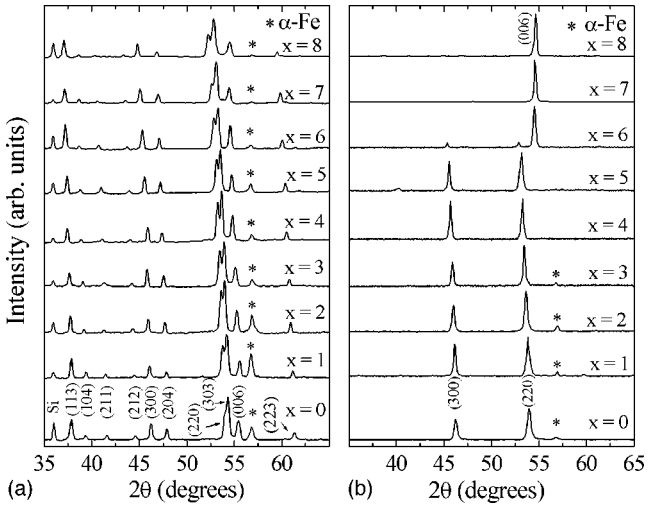

FIG. 1. X-ray diffraction patterns of $SmTbFe_{17-x}Ga_x$. (a) Free particles. (b) Field oriented particles.

TABLE I. Lattice parameters, unit cell volumes, and anisotropy fields H_A of $SmTbFe_{17-x}Ga_x$.

x	a(Å)	c(Å)	c/a	V(Å^3)	H_A(kOe)
0	8.535	12.480	1.462	787	32.9
1	8.560	12.490	1.459	793	29.3
2	8.594	12.521	1.457	800	26.9
3	8.608	12.547	1.458	805	24.6
4	8.639	12.625	1.461	816	22.2
5	8.660	12.640	1.459	821	18.2
6	8.703	12.675	1.456	831	8.5
7	8.736	12.676	1.451	838	15.2
8	8.791	12.658	1.440	847	16.1

expands with Ga, magnetization decreases. Such observations have been reported by others as well; Sabiryanov and Jaswal form the band structure calculations in Ga doped Sm_2Fe_{17}.[12] They have reported that substitution of Ga causes narrowing of the Fe 3*d* band due to reduced hybridization. On the other hand, the weakly polarized Ga states at the bottom of the Fe 3*d* band promote similar iron states mostly from the majority spin to this energy region. This effect decreases the magnetization. At higher Ga concentration, the magnetic dilution effect may take over the band modification effects.

The Curie temperature of $SmTbFe_{17-x}Ga_x$ increases from 408 K for $SmTbFe_{17}$ to 586 K for $x=3$ compound and decreases with further increase in Ga. The effect of Tb substitution is that the Curie temperatures are slightly less than those for the corresponding $Sm_2(Fe_{1-x}Ga_x)_{17}$.[13] Ga when substituted for Fe in R_2Fe_{17} systems occupies 18*h* site for low Ga concentrations.[14] This and the lattice expansion probably cause narrowing of the Fe 3*d* band. Sabiryanov and Jaswal[15] have shown that substitution of Ga in Sm_2Fe_{17} causes the 3*d* band to become narrower leading to an increase of the exchange interactions. Huang and Ching[16,17] have shown from the electronic structure calculations in Al/Ga/Si substituted Nd_2Fe_{17} that the variation in T_C mainly depends on the density of states (DOS) at E_F. They have obtained partial density of states of Ga (4*p*) band that has been hybridized with the Fe (3*d*) band and shown that the Ga (4*p*) band extends all the way to the Fermi level. Sabiryanov and Jaswal[15] have performed *ab initio* calculations of Curie temperature of $Sm_2Fe_{16}Ga$, by means of self-consistent spin polarized band structure calculation on the basis of a model proposed by Mohn and Wohlfarth[18] in terms of the DOS of the spin up and spin down bands at the Fermi level and have shown that a decrease in DOS at E_F enhances T_C. The increase in T_C seems to be related to the anisotropic expansion of the lattice with Ga. The c/a ratio is given in Table I. It decreases initially. This could be due to the preferential occupancy of Ga in the 18*h* sites, causing the expansion of a more than that of c. This may cause an effective band narrowing leading to the increase of T_C. However, the increase in c/a for $2 \leqslant x \leqslant 3$ could have been overwhelmed by the already narrowed 3*d* band (hence increase of T_C). Such an observation has been reported by Venkatesan *et al.*[19] as well in $Sm_2Fe_{17-x}Ga_x$. For higher Ga concentration in $SmTbFe_{17-x}Ga_x$ ($x>3$), 18*f* site too is occupied, causing the lattice to continue to expand. However, magnetic dilution may be predominant over the band narrowing, causing the T_C to decrease.

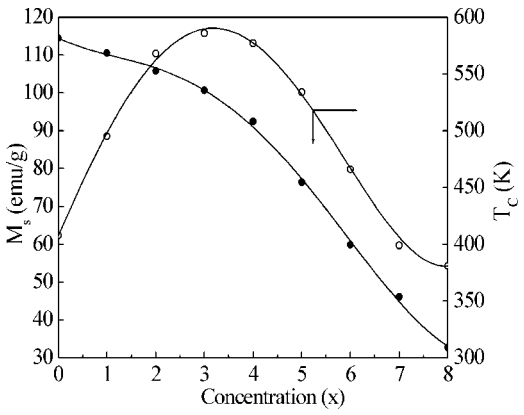

FIG. 2. Magnetization M_s and Curie temperature T_C of $SmTbFe_{17-x}Ga_x$.

The XRD patterns of magnetically aligned samples are shown in Fig. 1(b). Substitution of Ga is seen to change the easy magnetization direction (EMD) from planar to axial gradually from a Ga concentration of 6, as seen from a strong (006) reflection and the absence of any other reflections. In $Sm_2(Fe_{1-x}Ga_x)_{17}$, there is a region ($2 \leqslant x \leqslant 5$) in which the EMD is axial, whereas in $Tb_2(Fe_{1-x}Ga_x)_{17}$ the EMD is axial above $x=5$.[19,20] Thus, in the present case, Tb seems to have aided the stabilization of the uniaxial anisotropy. Probably, the presence of Tb modifies the crystalline electric fields (through the second order crystal field parameter A_2^0 coupled with negative α_J for Tb) aiding the EMD to remain axial. Magnetization measurements have been carried out at 80 K along the hard and easy directions. By extrapolating the magnetization curve along the hard direction to meet that along the easy direction, the anisotropy fields (H_A) have been determined. The variation of anisotropy field with the concentration is shown in Table I. The anisotropy field is found to decrease with x until $x \leqslant 6$ and starts increasing for the higher values of x.

IV. CONCLUSIONS

The substitution of Ga in $SmTbFe_{17}$ leads to an anisotropic expansion of lattice. The saturation magnetization decreases for the substitution of Ga substitutions and is attributed due to the change in the DOS at E_F. Curie temperature is found to increase for concentration up to 3 and to decrease

for higher concentrations, explained on the basis of band narrowing. The anisotropy is planar up to Ga concentration of 6 and becomes axial for higher concentrations. Tb is believed to stabilize the axial anisotropy.

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[1]J. M. D. Coey and H. Sun, J. Magn. Magn. Mater. **87**, L251 (1990).
[2]X-P. Zhong, R. J. Radwonski, F. R. de Boer, T. H. Jacob, and K. H. J. Buschow, J. Magn. Magn. Mater. **86**, 76 (1990).
[3]M. Valeaun, N. Plagaru, and E. Burgo, Solid State Commun. **89**, 519 (1994).
[4]B.-g. Shen, F. Wang, L. Kong, and L. Cao, J. Phys.: Condens. Matter **5**, 1685 (1993).
[5]T. H. Jacob, K. H. J. Buschow, G. F. Zhou, X. Li, and F. R. de Boer, J. Magn. Magn. Mater. **116**, 220 (1992).
[6]Z. W. Li, X. Z. Zhou, and A. H. Morrish, Phys. Rev. B **51**, 2891 (1995).
[7]K. G. Suresh and K. V. S. Rama Rao, J. Appl. Phys. **79**, 345 (1996).
[8]M. Venkatesan, U. V. Varadaraju, and K. V. S. Rama Rao, J. Magn. Magn. Mater. **182**, 231 (1998).
[9]K. G. Suresh and K. V. S. Rama Rao, IEEE Trans. Magn. **31**, 3722 (1995).
[10]J. C. Ingersoll, G. Markandeyulu, V. S. Murty, and K. V. S. Rama Rao, J. Alloys Compd. **354**, 29 (2003).
[11]K. V. S. Rama Rao, H. Ehernberg, G. Markandeyulu, U. V. Varadaraju, M. Venkatesan, K. G. Suresh, and V. S. Murthy, Phys. Status Solidi A **189**, 373 (2002).
[12]W. Z. Li, N. Tang, X. P. Zhong, J. L. Wang, Y. H. Gao, F. Yang, and Y. W. Zeng, J. Alloys Compd. **209**, 245 (1994).
[13]R. F. Sabiryanov and S. S. Jaswal, J. Appl. Phys. **81**, 5615 (1997).
[14]G. J. Long, G. K. Marasinghe, S. Mishra, O. A. Pringle, F. Gandjean, and K. H. J. Buschow, Solid State Commun. **88**, 761 (1993).
[15]R. F. Sabiryanov and S. S. Jaswal, J. Appl. Phys. **79**, 5942 (1996).
[16]M.-Z. Huang and W. Y. Ching, J. Appl. Phys. **76**, 7046 (1994).
[17]M.-Z. Huang and W. Y. Ching, J. Appl. Phys. **79**, 5546 (1996).
[18]P. Mohn and E. P. Wohlfarth, J. Phys. F: Met. Phys. **17**, 2421 (1987).
[19]M. Venkatesan, U. V. Varadaraju, and K. V. S. Rama Rao, Physica B **291**, 159 (2000).
[20]B.-g. Shen, *et al.*, Appl. Phys. Lett. **67**, 1621 (1995).