

## Effect of Al and Si substitutions on the magnetic properties of Sm Tb Fe 17

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**Effect of Al and Si substitutions on the magnetic properties of SmTbFe<sub>17</sub>**

J. C. Ingersoll, G. Markandeyulu,<sup>a)</sup> V. S. Murty, and K. V. S. Rama Rao  
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The structural and magnetic properties of SmTbFe<sub>17-x</sub>Al<sub>x</sub> ( $x=0-8$ ) and SmTbFe<sub>17-x</sub>Si<sub>x</sub> ( $x=0-3.5$ ) were investigated by x-ray-diffraction and magnetization studies. All the compounds stabilize in rhombohedral structure. The lattice parameters are found to increase with Al substitution whereas they are found to decrease with Si substitution. The rates of the decrease of saturation magnetization ( $M_s$ ) value with the substitution of Al and Si are almost the same. The easy direction of magnetization (EMD) is in the  $ab$  plane up to an Al concentration of  $x=7$  and has a tendency to shift towards the  $c$  axis at  $x=8$ . All the compounds with Si are seen to have the EMD in the  $ab$  plane. Electron-magnon scattering is seen to contribute to the electrical resistivity in both Al- and Si-substituted compounds; in the temperature range of 30–60 K and at higher temperatures, scattering due to phonons is observed. © 2005 American Institute of Physics.

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**INTRODUCTION**

Interstitially modified (with N or C) Sm<sub>2</sub>Fe<sub>17</sub> has been reported to have energy product at room temperature and Curie temperatures high enough to make it useful for permanent magnet applications.<sup>1,2</sup> In the R<sub>2</sub>Fe<sub>17</sub>N/C compounds, the Fe–Fe distances are more as compared to those in the parent compounds, leading to a substantial increase in their Curie temperatures ( $T_C$ ). However, they are thermodynamically unstable and decompose into rare-earth nitrides and  $\alpha$ -Fe at high temperatures. An alternative route to obtain increased bond distances, and hence Curie temperatures, is to force the lattice to expand by replacing some of the iron by either magnetic or nonmagnetic elements with larger metallic radii. Several R<sub>2</sub>Fe<sub>17-x</sub>M<sub>x</sub> ( $R=Ho, Y, Sm, Ce, Pr, \text{ and } Nd; M=Al, Ga, Si, \text{ and } Co$ ) compounds have been prepared<sup>3,4</sup> and their Curie temperatures have been found to be more than their respective parent compounds. Suresh and Rama Rao<sup>5</sup> have reported that the substitution of Al for Fe in ErPrFe<sub>17</sub> has caused the  $T_C$  to increase from 285 to 435 K for Al=3 and decrease with a further increase of Al. Venkatesan *et al.*<sup>6</sup> have reported an increase in  $T_C$  in HoErFe<sub>17-x</sub>Ga<sub>x</sub> with increasing concentration of Ga up to  $x=4$ . Ingersoll *et al.*<sup>7</sup> have reported that the substitution of Ga and Si for Fe in ErPrFe<sub>17</sub> has increased  $T_C$  until Ga and Si =3.5. In Sm<sub>2</sub>Fe<sub>17</sub> it has been found that the substitution of Ga or Al induces uniaxial magnetocrystalline anisotropy at room temperature<sup>8,9</sup> due to Sm having positive Stevens' factor ( $\alpha_j$ ) occupying the  $6c$  site having negative  $A_2^0$  (second-order crystalline electric-field parameter). In SmTbFe<sub>17</sub> compounds Ingersoll *et al.*<sup>10</sup> have reported an increase in the Curie temperature and the presence of axial anisotropy with Ga substitution. In this paper, the effects of the substitution of Al and Si on the structural, magnetic, and electrical properties of SmTbFe<sub>17</sub> compounds are presented.

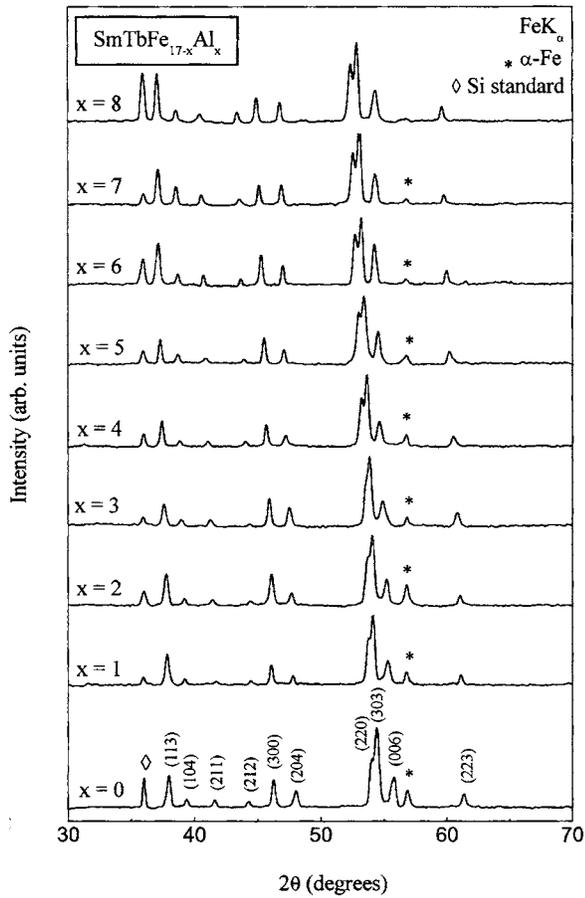
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**EXPERIMENTS**

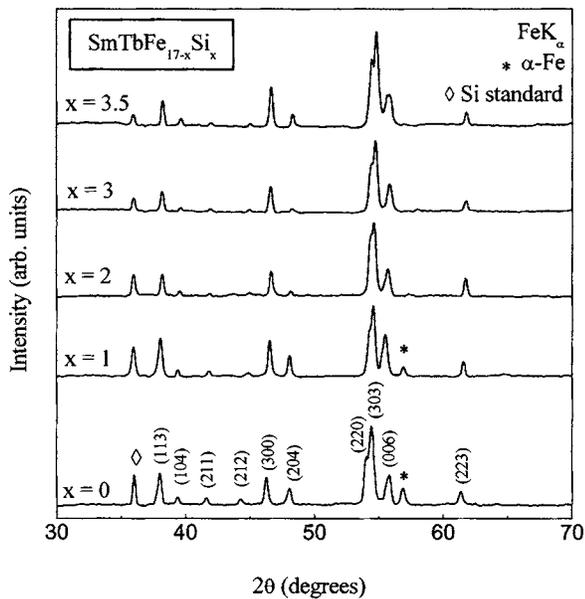
All the compounds were prepared by arc melting the stoichiometric amounts of high-purity elements (Sm, Tb: 99.9%; Fe: 99.95%; and Al, Si: 99.99%) in an argon atmosphere. Excess of Sm was taken to compensate for the weight loss during the melting. The ingots were melted several times to ensure the homogeneity. Subsequently the ingots were wrapped in tantalum foils, sealed in quartz tube in high-purity argon atmosphere at less than 100 torr pressure, annealed at 950 °C for 7 days, and quenched in the ice water mixture. X-ray-diffraction patterns on powder samples were taken employing Fe  $K\alpha$  radiation. Magnetization measurements were carried out using a vibrating-sample magnetometer (Model No. PAR 155) up to a field of 12 kOe and in the temperature range of 80–650 K. The saturation magnetization values were obtained from the Honda plots. Electrical resistivity measurements were carried out using four-probe method. Disk-shaped samples ( $\approx 6$  mm diameter and 1 mm thickness) were cut from the annealed ingots using a diamond cutter and polished. These samples were subsequently annealed at 1000 K for 24 h to remove the strains. A constant current source (Keithley 224) delivering a current of 100 mA and a nanovoltmeter (Keithley 181) were used for the measurements. Silver paint was used to establish the contacts in the temperature range of 30–300 K and pressure contacts were made for high-temperature (300–650 K) experiments.

**RESULTS AND DISCUSSION**

The powder x-ray-diffraction patterns for SmTbFe<sub>17-x</sub>Al<sub>x</sub> ( $x=0-8$ ) and SmTbFe<sub>17-x</sub>Si<sub>x</sub> ( $x=0-3.5$ ) compounds are shown in Figs. 1(a) and 1(b), respectively. All the compounds are found to have formed in the rhombohedral structure with traces of  $\alpha$ -Fe. However, the amount of  $\alpha$ -Fe phase is seen to be small at higher concentrations of



(a)



(b)

FIG. 1. (a) X-ray-diffraction patterns of  $\text{SmTbFe}_{17-x}\text{Al}_x$ . (b) X-ray-diffraction patterns of  $\text{SmTbFe}_{17-x}\text{Si}_x$ .

$x(x > 5)$ .  $\text{SmTbFe}_{17-x}\text{Si}_x$  compounds could be prepared to form in 2:17 phase up to  $x=3.5$  only in spite of the best efforts.

The substitution of Al for Fe results in the increase in the lattice parameters  $a$  and  $c$  and therefore the volume, as

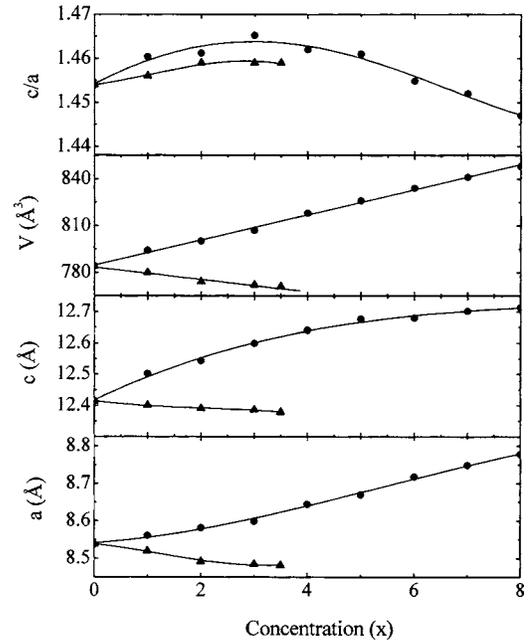


FIG. 2. Cell parameters and unit-cell volume and  $c/a$  in  $\text{SmTbFe}_{17-x}\text{Al}_x$  and  $\text{SmTbFe}_{17-x}\text{Si}_x$ .

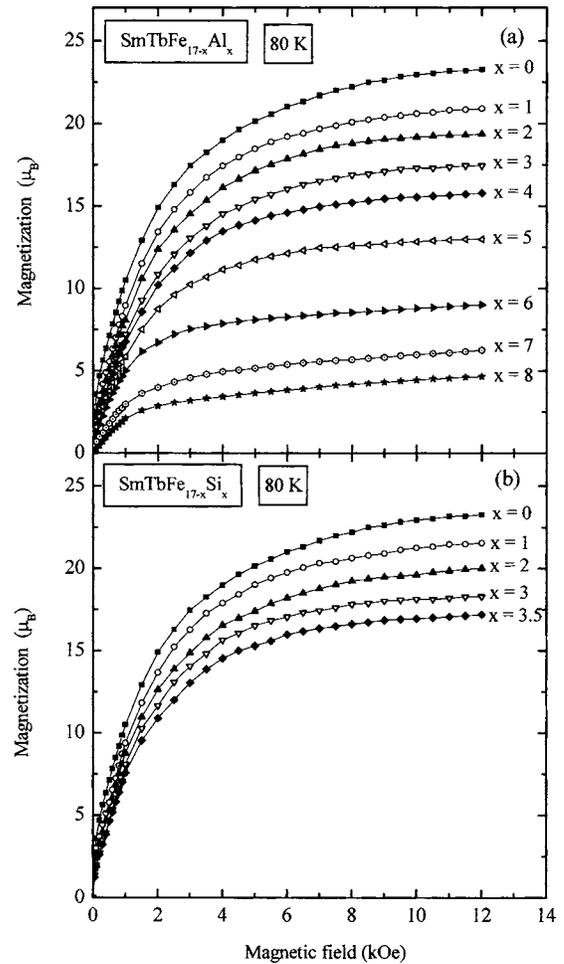


FIG. 3. Magnetization curves of (a)  $\text{SmTbFe}_{17-x}\text{Al}_x$  and (b)  $\text{SmTbFe}_{17-x}\text{Si}_x$ .

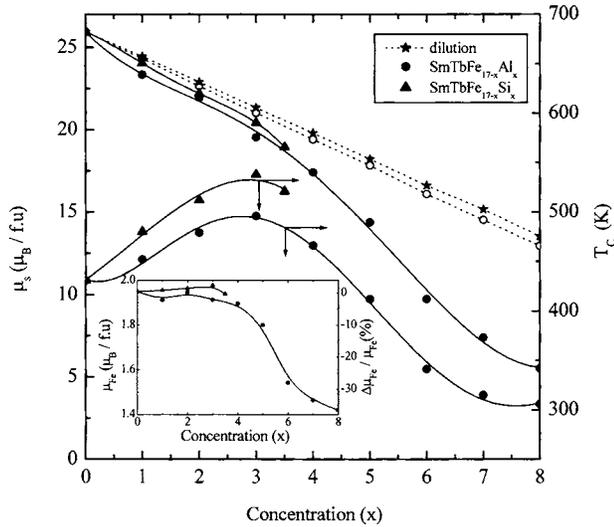


FIG. 4. Saturation magnetic moment ( $\mu_s$ ) and Curie temperature ( $T_C$ ) of  $\text{SmTbFe}_{17-x}\text{Al}_x$  and  $\text{SmTbFe}_{17-x}\text{Si}_x$ . The calculated  $\mu_s$  using the free-ion moments of Sm and Tb are shown with the symbol “○,” whereas the same using the reduced moments of Sm and Tb are shown with the symbol “★.” The fit lines are guides to the eye.

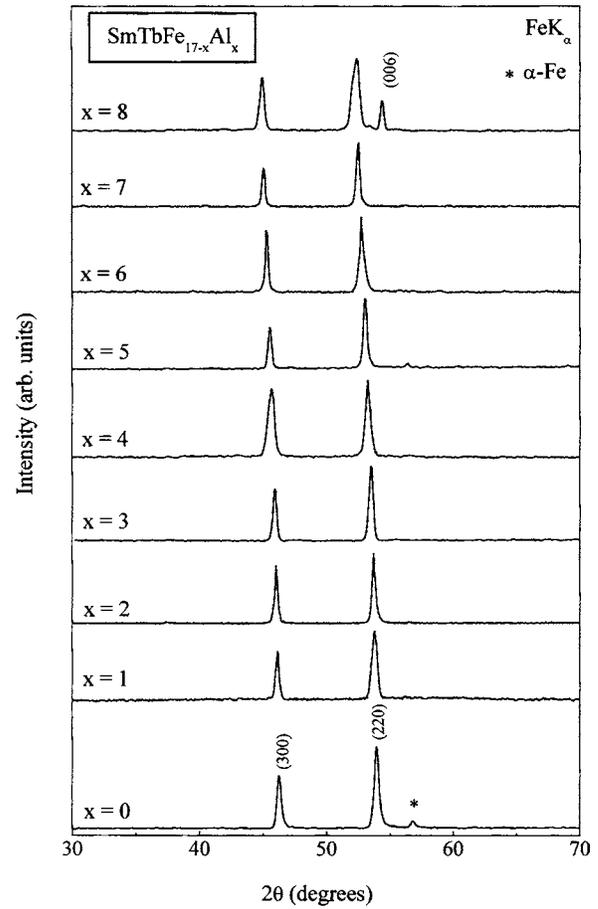
shown in Fig. 2. In  $\text{SmTbFe}_{17-x}\text{Al}_x$  compounds, the  $c/a$  value reaches a maximum at  $x=3$ , whereas the maximum  $c/a$  value for  $x=1$  remains the same until  $x=3.5$  in the case of  $\text{SmTbFe}_{17-x}\text{Si}_x$  compounds. The values and variation with  $x$  of  $c/a$  are similar to that reported<sup>11,12</sup> for  $\text{Nd}_2\text{Fe}_{17-x}\text{Al}_x$  and  $\text{Tb}_2\text{Fe}_{17-x}(\text{Ga}/\text{Si})_x$ , where Al/Ga/Si have been reported to occupy the  $18h$  site up to  $x=3$  and then  $18f$  and  $6c$  sites.

The magnetic-moment values in all the compounds have reached almost their respective saturation values, as shown in Figs. 3(a) and 3(b). The saturation magnetic moments ( $\mu_s$ ) at 80 K are shown in Fig. 4 for different Al and Si concentrations. The  $\mu_s$  value decreases from  $25.93\mu_B/\text{f.u.}$  for  $x=0$  to  $5.52\mu_B/\text{f.u.}$  for  $x=8$  in  $\text{SmTbFe}_{17-x}\text{Al}_x$  compounds while it decreases to  $18.93\mu_B/\text{f.u.}$  for  $x=3.5$  in  $\text{SmTbFe}_{17-x}\text{Si}_x$  compounds. The rates of the decrease of saturation magnetization values with  $x$  are almost the same in both the series up to  $x=3$ .

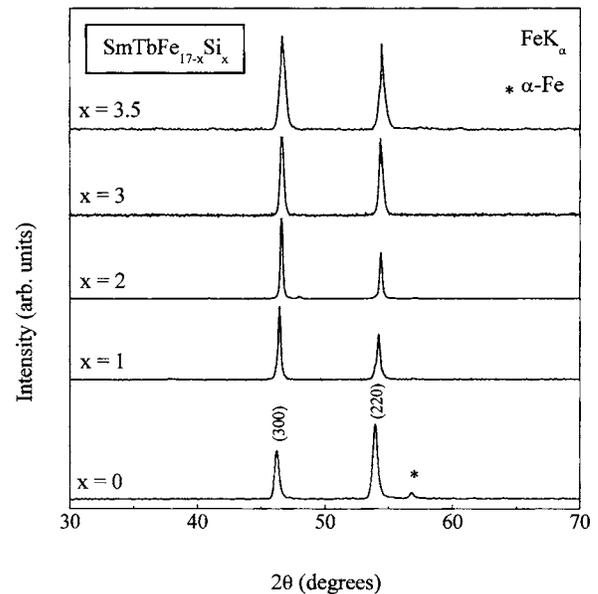
In the molecular-field description of the magnetic moments of  $R_2T_{17-x}M_x$ ,  $\mu_s$  has been given as<sup>13</sup>

$$\mu_s = (17-x)\mu_{\text{Fe}} + \mu_{\text{Sm}} - \mu_{\text{Tb}}, \quad (1)$$

where  $\mu_{\text{Fe}}$  is the magnetic moment of Fe and  $\mu_{\text{Sm}}$  and  $\mu_{\text{Tb}}$  are the magnetic moments of Sm and Tb, respectively. It is known that the Sm moments are coupled ferromagnetically and the Tb moments are coupled antiferromagnetically to Fe moments.<sup>14,15</sup> For  $x=0$  compound,  $\mu_{\text{Fe}}$  has been calculated using the experimental  $\mu_s$  value and the moments of Sm and Tb (reduced with respect to the respective free-ion moments) are taken from those obtained from the mean-field analysis of  $\text{Sm}_{1-x}\text{Tb}_x\text{Fe}_{17}$  (Ref. 16) and is found to be  $1.95\mu_B$ . Using this value,  $\mu_s$  for Al- and Si-substituted  $\text{SmTbFe}_{17}$  were calculated assuming that Al and Si contribute only to the dilution of Fe moments and the values are also shown in Fig. 4. The above exercise was also done by assuming the free-ion moments on Sm and Tb and the results are also shown in Fig. 4. It is seen that the variation in the rare-earth moment



(a)



(b)

FIG. 5. (a) X-ray-diffraction patterns of  $\text{SmTbFe}_{17-x}\text{Al}_x$  powders aligned in an external magnetic field. (b) X-ray-diffraction patterns of  $\text{SmTbFe}_{17-x}\text{Si}_x$  powders aligned in an external magnetic field.

has an insignificant impact on the moments of Fe. Comparing these values with the experimental  $\mu_s$  values which are smaller, it is seen that the variation in the magnetic moment with Al/Si substitution has, in addition to the dilution effect, the contribution from the charge-transfer effect due to the

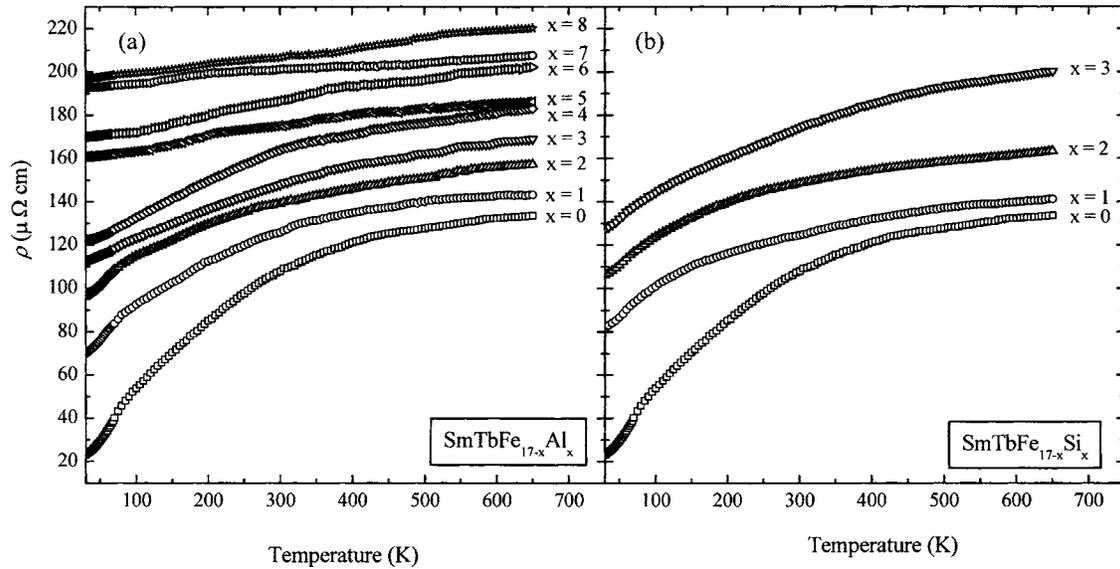


FIG. 6. Temperature variation of the electrical resistivity of (a)  $\text{SmTbFe}_{17-x}\text{Al}_x$  and (b)  $\text{SmTbFe}_{17-x}\text{Si}_x$ .

$4p$ - $3d$  hybridization. The net  $\mu_{\text{Fe}}$  values were calculated for all the compounds from the experimental  $\mu_s$  values using the reduced moments for Sm and Tb (Ref. 16) and are shown in the inset of Fig. 4. The value of  $\mu_{\text{Fe}}$  is found to vary from  $1.95\mu_B/\text{f.u.}$  for  $x=0$  to  $1.42\mu_B/\text{f.u.}$  for  $x=8$  in the case of  $\text{SmTbFe}_{17-x}\text{Al}_x$  and to  $1.94\mu_B/\text{f.u.}$  for  $x=3.5$  in the case of  $\text{SmTbFe}_{17-x}\text{Si}_x$ .

Huang and Ching,<sup>17,18</sup> from the band-structure calculations in  $\text{Nd}_2\text{Fe}_{17}$  with Ga, Si, and Al substitutions, have reported a hybridization of orbitals of Fe with the  $3p$  orbitals of Al and Ga and the  $4p$  orbitals of Si and obtained their partial density of states. Also, they have reported that the weakly polarized Si and Al states at the bottom of the Fe  $3d$  band promote similar iron states mostly from the majority spin to this energy region leading to a decrease in the magnetization of Fe sublattice and hence the magnetization.

The  $T_C$  of  $\text{SmTbFe}_{17-x}\text{Al}_x$  compounds is seen to increase from 431 K for  $x=0$  to 496 K for  $x=3$  and decreases substantially. In the case of  $\text{SmTbFe}_{17-x}\text{Si}_x$  compounds, the  $T_C$  reaches a maximum value of 538 K for  $x=3$  and decreases for a further increase of  $x$  (Fig. 4). Jacobs *et al.*<sup>19</sup> have attributed such an increase in  $T_C$  to the volume expansion. Interstitial modification by N or C has also been reported to expand the lattice and subsequently increase in the magnetization and Curie temperature as well due to band narrowing.<sup>5,6</sup> However, in the case of Si-substituted  $\text{SmTbFe}_{17}$  compounds, it has been observed that  $T_C$  increases while the unit-cell volume decreases. Probably the broader Si band at the bottom of  $3d$  band<sup>18</sup> causes charge transfer sufficient to cause an increase in the  $T_C$ .

It is observed that the increase in the Curie temperature in Si-substituted compounds is more than that in the Al-substituted compounds. Sabiryanov and Jaswal<sup>20</sup> have done *ab initio* calculations of the Curie temperature of  $\text{Sm}_2\text{Fe}_{16}M$ , where  $M$  is Ga, Al, and Si by means of the self-consistent spin-polarized band-structure calculation on the basis of a model proposed by Mohn and Wohlfarth.<sup>21</sup> In this model

$$T_C \propto \frac{M_0}{\chi_0}, \quad (2)$$

where  $M_0$  is the Fe magnetization at 0 K per unit cell and  $\chi_0$  is the exchange-enhanced ferromagnetic susceptibility, at 0 K, and is given by

$$\chi_0^{-1} = \left[ \frac{1}{2N^\uparrow(E_F)} + \frac{1}{N^\downarrow(E_F)} - 1 \right] \frac{1}{2\mu_B^2}, \quad (3)$$

where  $N^\uparrow(E_F)$  and  $N^\downarrow(E_F)$  are, respectively, the average spin-up and spin-down densities of states per Fe atom at the Fermi level and  $I$  is the Stoner parameter. The changes in the  $T_C$  depend strongly on the changes in  $N^\uparrow(E_F)$  and  $N^\downarrow(E_F)$ . In general, a decrease in density of states (DOS) at  $E_F$  enhances  $T_C$  as can be seen from relations (2) and (3). Thus in Si-substituted compounds, the changes may be more than in the Al-substituted compounds due to the larger width of the partial density of states (PDOS) of Si.

The x-ray-diffraction patterns of magnetically aligned (perpendicular to the substrate) samples of  $\text{SmTbFe}_{17-x}\text{Al}_x$  and  $\text{SmTbFe}_{17-x}\text{Si}_x$  compounds are shown in Figs. 5(a) and 5(b). In Al-substituted  $\text{SmTbFe}_{17}$  compounds planar anisotropy exists up to  $x=7$ , as seen by the presence of (300) and (220) peaks and the absence of any other peak. In the  $\text{SmTbFe}_9\text{Al}_8$  compounds, (006) peak, in addition to (300) and (220) peaks, is seen indicating a tendency for the anisotropy to shift from planar to axial [Fig. 5(a)]. It can be clearly seen from Fig. 5(b) that in all the Si-substituted compounds, the easy magnetization direction is planar in the concentration range investigated.

## ELECTRICAL RESISTIVITY

The temperature variation of the electrical resistivity of  $\text{SmTbFe}_{17-x}\text{Al}_x$  ( $x=0-7$ ) and  $\text{SmTbFe}_{17-x}\text{Si}_x$  ( $x=0-3$ ) compounds is shown in Figs. 6(a) and 6(b), respectively. The resistivity increases as the concentration of Al/Si is increased in the entire temperature range. The resistivity of the

TABLE I. Parameters for  $\text{SmTbFe}_{17-x}\text{Al}_x$  compounds after the resistivity data were fitted to the equations  $\rho(T)=\rho_0+AT^2$  (30–60 K) and  $\rho(T)=B+CT-DT^3$  (70–350 K).

$x$	$\rho$ ( $\mu\Omega$ cm)	$A$ ( $\text{n}\Omega$ cm $\text{K}^{-2}$ )	$B$ ( $\mu\Omega$ cm)	$C$ ( $\mu\Omega$ cm $\text{K}^{-1}$ )	$D \times 10^{-7}$ ( $\mu\Omega$ cm $\text{K}^{-3}$ )
0	19.11	4.18	18.29	0.36	6.57
1	66.38	3.90	71.53	0.21	3.67
2	93.27	3.42	99.62	0.16	2.89
3	111.79	1.38	109.20	0.14	1.48
4	121.00	1.17	112.30	0.30	3.40
5	160.45	0.37	156.05	0.07	0.96
6	169.46	0.39	164.25	0.08	0.31
7	192.40	0.28	190.65	0.04	0.96
8	196.50	0.40	194.80	0.04	0.49

silicon compounds is higher at all temperatures than that of the corresponding Al-substituted compounds. This presumably occurs because of the reduced number of conduction electrons in the silicon compounds, a reduction, which occurs as a result of the higher covalency of the silicon bonds with its near neighbors as compared to the aluminum bonds.

The total electrical resistivity of a ferromagnetic material according to Matthiessen's rule<sup>22</sup> is expressed as

$$\rho(T) = \rho_0 + \rho_{\text{ph}}(T) + \rho_{\text{mag}}(T), \quad (4)$$

where  $\rho_0$  is the temperature-independent residual resistivity,  $\rho_{\text{ph}}(T)$  is the contribution due to electron-phonon scattering, and  $\rho_{\text{mag}}(T)$  is the electron-magnon scattering. One of these contributions predominates depending upon the temperature range.

At low temperature the electrical resistivity in these compounds is attributed to electron-electron scattering and electron-magnon scattering, which has been shown to have a quadratic dependence with temperature in the temperature range of 30–60 K. Thus, the temperature dependences of the electrical resistivity in  $\text{SmTbFe}_{17-x}\text{Al}_x$  and  $\text{SmTbFe}_{17-x}\text{Si}_x$  are fitted to

$$\rho(T) = \rho_0 + AT^2. \quad (5)$$

The values of  $A$  obtained from the fit are given in Tables I and II. The values are seen to be of the order of  $\text{n}\Omega$  cm  $\text{K}^{-2}$  indicating the dominance of the electron-magnon contribution over the electron-electron contribution to the electrical resistivity.<sup>23</sup> There is a large change in the value of  $A$  for  $\text{SmTbFe}_{14}\text{Al}_3$  from that of the case of  $\text{SmTbFe}_{15}\text{Al}_2$ . In several  $R_2\text{Fe}_{17-x}\text{Al}_x$  compounds it has been reported that until  $x \approx 3$ , Al occupies the 18*h* site and starts occupying the 18*f*

TABLE II. Parameters for  $\text{SmTbFe}_{17-x}\text{Si}_x$  compounds after the resistivity data were fitted to the equations  $\rho(T)=\rho_0+AT^2$  (30–60 K) and  $\rho(T)=B+CT-DT^3$  (70–350 K).

$x$	$\rho$ ( $\mu\Omega$ cm)	$A$ ( $\text{n}\Omega$ cm $\text{K}^{-2}$ )	$B$ ( $\mu\Omega$ cm)	$C$ ( $\mu\Omega$ cm $\text{K}^{-1}$ )	$D \times 10^{-7}$ ( $\mu\Omega$ cm $\text{K}^{-3}$ )
0	19.11	4.18	18.29	0.36	6.57
1	79.14	3.05	91.03	0.130	1.83
2	103.56	2.90	110.45	0.150	3.00
3	124.63	2.90	129.40	0.150	1.26

site above  $x=3$ . This could probably lead to the development of shorter-wavelength magnons that could result in the decrease in the curvature of the parabolic part of  $\rho(T)$  curve, reflecting as smaller  $A$  values.

At higher concentrations the temperature coefficient of the resistivity is considerably reduced. This may be because of the small mean free paths of the charge carriers as a result of the increase in the scattering. In the case of Si-substituted  $\text{SmTbFe}_{17}$  compounds the electron-magnon contribution is observed for the concentrations  $x=0$  and 1 as the temperature-dependent electrical resistivity shows a quadratic variation in the temperature range of 30–60 K.

A fit of the resistivity of  $\text{SmTbFe}_{17-x}\text{Al}_x$  and  $\text{SmTbFe}_{17-x}\text{Si}_x$  compounds  $x$ , between 120 and 295 K, to the function,

$$B + CT^2 - DT^3 \quad (6)$$

indicates that  $D$  is positive and equal to  $\approx 3 \mu\Omega$  cm  $\text{K}^{-3}$ , for  $x=1$  and  $x=3$ , for instance, for both Al and Si substitutions (see Tables I and II). This value is similar to those obtained<sup>24</sup> in  $(\text{Pr}_x\text{Er}_{1-x})_2\text{Fe}_{17}$  compounds. A positive  $D$  value implies that the quantity  $[3(1/N)^2(dN/d\varepsilon)^2 - (1/N)(d^2N/d\varepsilon^2)]$  is positive, which is possible if the Fermi level is situated in the flank of the density-of-states valley, where the slope  $(dN/d\varepsilon)$  is large and the curvature  $(d^2N/d\varepsilon^2)$  is small. Band-structure calculations<sup>18</sup> on the  $\text{Nd}_2\text{Fe}_{17-x}(\text{Al}/\text{Si})_x$  solid solutions have shown a similar result for the Fermi level.

## SUMMARY AND CONCLUSIONS

The substitution of Al leads to an expansion of lattice whereas it decreases due to the substitution of Si for Fe in  $\text{SmTbFe}_{17}$ . The saturation magnetization decreases with the increase of the amount of Al and Si substitutions. The rates of the decrease of  $M_s$  are similar in the case of Al- and Si-substituted compounds and are attributed to the changes in the density of states of 3*d* band at the Fermi level. The Curie temperature increases initially with Al and Si substitutions and then decreases. The easy magnetization direction lies on the basal plane up to  $x=7$  in  $\text{SmTbFe}_{17-x}\text{Al}_x$  and for the  $x=8$  compound there is a tendency for the easy direction of magnetization (EMD) to move towards  $c$  axis. In  $\text{SmTbFe}_{17-x}\text{Si}_x$  the EMD is planar up to  $x=3.5$ . The electron-magnon scattering contribution to the electrical resistivity is observed at the temperature range of 30–60 K in all the compounds.

## ACKNOWLEDGMENT

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