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Influence of Si and Co substitutions on magnetoelastic properties of R_2Fe_{17} (R = Y, Er and Tm) intermetallic compounds

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ABSTRACT

The magnetostriction of the off-stoichiometric R2Fe17-type intermetallic compounds based on $R_2Fe_{14-x}Co_xSi_2$ (R = Y, Er, Tm and x = 0, 4) was measured, using the strain gauge method in the temperature range 77-460 K under applied magnetic fields up to 1.5 T. All compounds show sign change and reduction in magnetostriction values compared to the R₂Fe₁₇ compounds by Si substitution. For $Y_2Fe_{14}Si_2$ and $Er_2Fe_{14}Si_2$, saturation behaviour is observed near magnetic ordering temperature (T_c), whereas for $Tm_2Fe_{14}Si_2$, saturation starts from T > 143 K. Also, Co substitution has different effects on the magnetostriction of R₂Fe₁₄Si₂ compounds. In Er₂Fe₁₀Co₄Si₂ and Tm₂Fe₁₀Co₄Si₂, saturation occurs below the spin reorientation temperature (T_{SR}). In addition, in $Er_2Fe_{14}Si_2$, a sign change occurs in the anisotropic magnetostriction ($\Delta\lambda$) as well as the volume magnetostriction ($\Delta V/V$) at their T_{SR} values. The volume magnetostrictions of the Tm-containing compounds show an anomaly around their T_{SR} . In $R_2Fe_{14}Si_2$ compounds, parastrictive behaviour is also observed in $\Delta V/V$ near their T_C values. In addition, the magnetostriction of the sublattices is investigated. Results show that in $R_2Fe_{14}Si_2$ compounds, the rare-earth sublattice contribution to magnetostriction is negative and comparable to the iron sublattice, whereas, in R₂Fe₁₀Co₄Si₂ compounds, the rare-earth sublattice contribution is positive and larger than Fe sublattice. These results are discussed based on the effect of Si and Co substitutions on the anisotropy field of these compounds. Influence of the spin reorientation transition on the magnetostriction of these compounds is discussed in terms of the anisotropic sublattice interactions.

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1. Introduction

Intermetallic compounds based on rare-earth (R) and transition-metal (T) elements have an important role from technological and fundamental points of view. The low Curie temperature and easy plane anisotropy of R_2Fe_{17} compounds confines their applications as permanent magnets [1–3]. In the past decade, many investigations were performed on R_2Fe_{17} compounds with the aim of improving their magnetic properties for practical applications as permanent magnets. For example, partial substitution of Cr, Si, Ga and Al for iron atoms in several R_2Fe_{17} compounds leads to significant increase in Curie temperature and to modifying their magnetic anisotropy [4–9].

 R_2Fe_{17} compounds are either ferromagnetic system with R = light rare-earth or ferrimagnetic system with R = heavy rare-earth element [10,11]. Our $R_2Fe_{14-x}Co_xSi_2$ compounds crystallize in the hexagonal structure of Th_2Ni_{17} -type with P6₃/mmc space group, where the R ions are located on two different crytallogrophic sites (2b and 2d) and the T atoms occupy four non-

equivalent sites (4f, 6g, 12j and 12k) [10–12]. Previous results on these compounds showed that the lattice parameters decrease and Curie temperatures increase for Si and Co substitutions for Fe [10,11]. Also, partial substitution of Si decreases the saturation magnetisation (M_s). Recently, investigations using Mössbauer spectroscopy on several R₂Fe₁₄Si₂ and R₂Fe₁₀Co₄Si₂ off-stoichiometric compounds showed that the values of the magnetic hyperfine fields corresponding to the four non-equivalent lattice sites increase after Co substitution [13,14]. This is consistent with the increase of M_s by Co substitution for Fe in the presence of Si [11].

The rare-earth (R = Er, Tm) sublattice in these compounds has competitive magnetic anisotropy with Fe-sublattice anisotropy. Substitution of Si enhances the contribution of the negative second-order crystal field parameter (A_2^0) [5,6], so that the R sublattice anisotropy is strengthened in the R₂Fe_{17-x}Si_x compounds whenever R is a rare-earth element with positive Stevens factor (i.e. $\alpha_j > 0$), such as Er and Tm. Hence, partial replacement of Fe by Si induces a spin reorientation transition in Er₂Fe₁₄Si₂ at 100 K and increases T_{SR} in Tm₂Fe₁₄Si₂ from 72 to 180 K. In addition, partial substitution of Fe by Co in both compounds leads to a shift in T_{SR} toward higher temperatures of 230 and 270 K, respectively [10,11]. The region of uniaxial anisotropy dominates in temperatures below the spin reorientation temperature (T_{SR}) of each compound. But, for

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Fig. 1. Temperature dependence of AC susceptibility for: (a) $Er_2Fe_{10}Co_4Si_2$ and (b) $Tm_2Fe_{10}Co_4Si_2$ compounds.

the Y_2Fe_{17} compound where Y is non-magnetic and the Fe sublattice favors easy plane anisotropy, substitutions of Si and Co for Fe do not modify the planar anisotropy, but weaken the anisotropy field [1,2,10,11]. Most of magnetic properties of R_2Fe_{17} compounds are influenced by Si and Co substitutions. Hence, in these compounds we expect the magnetoelastic interactions to be modified by Si and Co substitutions as well.

Some investigations have been performed on magnetoelastic properties of R_2Fe_{17} and their nitrides and hydrides [14–20]. However, so far no attention has been paid to the effect of non-magnetic elements (Si) in the presence of Co atoms on magnetostriction of these compounds until now.

In the present work, using the magnetostriction measurement method we have studied the contribution of rare-earth and transition-metal sublattices as well as the role of spin reorientation transition on the magnetoelastic behaviour of $R_2Fe_{14-x}Co_xSi_2$ (R = Y, Er and Tm) compounds.

2. Experimental methods

The starting materials were prepared by induction melting of the constituent metals in a water-cooled copper boat under a flowing argon atmosphere. As-cast ingots were heat-treated at 900 °C for 2 days and then rapidly cooled to room temperature. X-ray diffraction



Fig. 2. Field dependence of the anisotropic magnetostriction of: (a) $Y_2Fe_{14}Si_2$ and (b) $Y_2Fe_{10}Co_4Si_2$ compounds at selected temperatures (in this and all the following figures, the lines are guides for the eyes).

(Cu-K α radiation) patterns of the annealed samples revealed the formation of single-phase materials with the expected hexagonal crystal structure.

The AC-magnetic susceptibility of the Er and Tm-containing samples was measured between 77 and 300 K using Lake-Shore AC susceptometer model 7000 in fields lower than 80 A/m at a frequency of 111 Hz. The magnetostriction measurements were performed by the strain gauge method on disk-shaped bulk samples (10 mm diameter, 3 mm thick) in applied fields up to 1.5 T, over temperatures ranging from 77 to 460 K. The magnetostriction parameters were measured (with the accuracy of 2×10^{-6}) parallel (longitudinal magnetostriction, λ_1) and perpendicular (transverse magnetostriction λ_t) to the field direction, thus allowing deduction of the anisotropic magnetostriction ($\Delta \lambda = \lambda_1 - \lambda_t$) and the volume magnetostriction ($\Delta V/V = \lambda_1 + 2\lambda_t$).

3. Results and discussion

3.1. AC susceptibility

Fig. 1. shows temperature dependence of the AC susceptibility for the $Er_2Fe_{10}Co_4Si_2$ and $Tm_2Fe_{10}Co_4Si_2$ compounds. A cusp or

peak in each of the $\chi_{ac}(T)$ or $d\chi_{ac}/dT$ curves clearly indicates the spin reorientation transition temperature (T_{SR}). The observed values of T_{SR} for $Er_2Fe_{14}Si_2$ and $Er_2Fe_{10}Co_4Si_2$ are 100 and 230 K, and for $Tm_2Fe_{14}Si_2$ and $Tm_2Fe_{10}Co_4Si_2$ are 183 and 279 K, respectively. These results are consistent with the previous measurements determined for these compounds [10,11]

3.2. Magnetostriction of $Y_2Fe_{14-x}Co_xSi_2(x = 0 \text{ and } 4)$

Fig. 2 shows the anisotropic magnetostriction $(\Delta\lambda)$ of Y₂Fe₁₄Si₂ and Y₂Fe₁₀Co₄Si₂ as a function of applied field at selected temperatures. It is clear that saturation behaviour starts approximately from T = 378 and 207 K for compounds with x = 0 and 4, respectively. The difference between the values of $\Delta\lambda$ at T = 405 and 442 K for the sample with x = 0 is larger than the corresponding value in the compound with x = 4. This effect is due to the anisotropy field weakness close to the magnetic ordering temperature ($T_C = 465$ K) of the compound with x = 0[10].

The temperature dependence of $\Delta\lambda$ for these compounds at selected fields is shown in Fig. 3. The behaviour of the $\Delta\lambda$ curve of the sample with x = 0 is found to be different than for the compound with x = 4 at all applied magnetic fields. The result for $\Delta\lambda$ of the compound with x = 4 may indicate that the anisotropy

field of this compound is reduced compared to the corresponding value for the compound with x = 0 at all temperatures.

On the other hand, comparison of $\Delta\lambda$ curves for Y₂Fe₁₄Si₂ in Fig. 3a with the corresponding curve for Y₂Fe₁₇ in Ref. [17] shows opposite curvature and sign change in our compound. In addition, the absolute value of $\Delta\lambda$ at room temperature is increased at least by one order of magnitude by Si substitution. This is due to the increase of the Curie temperature from 310 K in Y₂Fe₁₇ [1] to 465 K in Y₂Fe₁₄Si₂ [10].

3.3. Magnetostriction of $Er_2Fe_{14-x}Co_xSi_2(x = 0 \text{ and } 4)$

For both compounds (x = 0 and 4), isothermal curves of the anisotropic magnetostriction ($\Delta\lambda$) as measured at selected temperatures are shown in Fig. 4. The anisotropic magnetostriction of $\text{Er}_2\text{Fe}_{14}\text{Si}_2$ exhibits a sign change at spin reorientation temperature (T_{SR}), whereas this phenomenon is not observed in $\text{Er}_2\text{Fe}_{10}\text{Co}_4\text{Si}_2$ and $\text{Er}_2\text{Fe}_{17}$ [16]. Below $T_{\text{SR}}(=100 \text{ K})$, the anisotropic magnetostriction becomes negative at low applied fields, and then monotonically increases and changes its sign at higher applied fields. With increasing temperature, $\Delta\lambda$ increases gradually and approaches saturation beyond T = 380 K. Also, magnetostriction compensation occurs in $\text{Er}_2\text{Fe}_{14}\text{Si}_2$ in a temperature range around its T_{SR} at different applied magnetic



Fig. 3. Temperature dependence of the anisotropic magnetostriction of: (a) Y₂Fe₁₄Si₂ and (b) Y₂Fe₁₀Co₄Si₂ compounds at selected magnetic fields.



Fig. 4. Field dependence of the anisotropic magnetostriction of: (a) $Er_2Fe_{14}Si_2$ and (b) $Er_2Fe_{10}Co_4Si_2$ compounds at selected temperatures.

fields. This may attribute to the sign change in the magnetoelastic coupling constant above T_{SR} .

From the comparison of magnetic ordering and spin reorientation temperatures of $\text{Er}_2\text{Fe}_{14}\text{Si}_2$ and $\text{Er}_2\text{Fe}_{14}\text{Si}_3$ compounds in Refs. [10,21], it can be concluded that the value of the anisotropy field of our off-stoichiometric sample below T_{SR} is about 0.5 T. Therefore, the inflection point on isothermal curves of $\Delta\lambda$ (in Fig. 4a) can be designated as the anisotropy field (H_A) of this compound. The low H region ($H < H_A$) has a crystal electric field origin corresponding to the rotation of the magnetisation from the *c*-axis to the applied field direction. The second region ($H > H_A$) is the forced magnetostriction, where the magnetisation vector is along the field.

From the $\Delta\lambda$ curves for Er₂Fe₁₀Co₄Si₂ (in Fig. 4b) it is clear that the anisotropic magnetostriction has a maximum value around $T_{\text{SR}}(\approx 230 \text{ K})$, and saturation behaviour takes place at low applied fields ($\approx 0.5 \text{ T}$) due to the weakness of the magnetocrystalline anisotropy.

The volume magnetostriction $(\Delta V/V)$ of the Er₂Fe_{14-x}Co_xSi₂ compounds are plotted versus applied field at selected temperatures in Fig. 5. It can be noted that the dependence of $\Delta V/V$ for Er₂Fe₁₄Si₂ (in Fig. 5a) show opposite behaviour to that of Er₂Fe₁₀Co₄Si₂ (in Fig. 5b) below and above the spin reorientation temperature ($T_{SR} \approx 100$ and 230 K for x = 0 and 4, respectively).



Fig. 5. Field dependence of the volume magnetostriction of: (a) $Er_2Fe_{14}Si_2$ and (b) $Er_2Fe_{10}Co_4Si_2$ compounds at selected temperatures.

The volume magnetostriction of the compound with x = 0 has negative (positive for x = 4) values below T_{SR} and positive (negative for x = 4) values above T_{SR} which indicates an expansion (contraction for x = 4) of the compound around T_{SR} . This result may be attributed to the different values of volumerelated magnetostrictive coupling constants. In $Er_2Fe_{10}Co_4Si_2$ beyond T_{SR} , the value of $\Delta V/V$ decreases at low applied fields, reaches a minimum, then monotonically increases. The fall of volume magnetostriction can be due to enhancement of the planar anisotropy field.

The volume magnetostriction curves of $\text{Er}_2\text{Fe}_{14}\text{Si}_2$ (in Fig. 5a) are similar to the isothermal curves of the anisotropic magnetostriction of this compound shown in Fig. 4a. However, for temperatures near T_C the $\Delta V/V$ curves tend to have a quadratic dependence on applied field as expected for the parastrictive behaviour. A similar behaviour was also observed in Y- and Tm-containing compounds.

3.4. Magnetostriction of $Tm_2Fe_{14-x}Co_xSi_2(x = 0 \text{ and } 4)$

Fig. 6 shows typical isothermal curves for the anisotropic magnetostriction of the studied samples. It is clear that the saturation behaviour occurs in both compounds. This behaviour in the compound with x = 0 starts at a lower temperature ($T \approx 143$ K



Fig. 6. Field dependence of the anisotropic magnetostriction of: (a) $Tm_2Fe_{14}Si_2$ and (b) $Tm_2Fe_{10}Co_4Si_2$ compounds at selected temperatures.

in Fig. 6a) than in the sample with x = 4 ($T \approx 187$ K in Fig. 6b), which is due to the lower spin reorientation temperature ($T_{SR} \approx 183$ and 279 K for x = 0 and 4, respectively) [11]. In Tm₂Fe₁₇, there is a sign change in the $\Delta\lambda$ isothermal curve when passing through the spin reoreintation temperature at 90 K [18]. But, such an effect is not observed in the Tm₂Fe₁₄Si₂ sample. This indicates that partial substitution of Si for Fe has strong effect on the overall magnetostrictions modes.

The temperature dependence of the anisotropic magnetostriction of both compounds at selected applied fields is presented in Fig. 7. For both samples at all applied fields, $\Delta\lambda$ increases up to a temperature designated as T_{max} . It is clear that T_{max} is much lower in the cobalt-free compound. This implies that the axial anisotropy field increases with Co substitution at low temperatures and this is due to the fact that Co induces axial anisotropy in these typical compounds [22–24]. Therefore, we need stronger applied fields to observe a pronounced maximum at $\Delta\lambda$ curve.

Fig. 8 shows the temperature dependence of the volume magnetostriction ($\Delta V/V$) for these compounds at selected applied fields. Both samples exhibit a cusp close to their spin reorientation transitions temperatures. These anomalies around T_{SR} can be ascribed to the decrease of the magnetic anisotropy and the rotation of magnetic moments along the applied field in the



Fig. 7. Temperature dependence of the anisotropic magnetostriction of $Tm_2Fe_{14}Si_2$ (open symbols) and $Tm_2Fe_{10}Co_4Si_2$ (filled symbols) compounds at selected magnetic fields.

corresponding temperature ranges. It is clear that $\Delta V/V$ increases once again with temperature beyond 225 and 375 K for x = 0 and 4, respectively. This indicates that the planar anisotropy field begins to decrease with increasing temperature. In addition, comparison of the $\Delta V/V$ values near T_{SR} in both compounds shows that the peak of the volume magnetostriction in the cobalt-free sample in the maximum applied field is approximately five times larger than in the other sample. It is well known that the contribution to the magnetic interaction between the localized moments. Therefore, reduction in the volume magnetostriction implies that magnetic interaction has been weakened by Co substitution in $Tm_2Fe_{14-x}Co_xSi_2$. This is quite similar to the observed behaviour in $Er_2Fe_{14-x}Co_xB$ compounds [25].

3.5. Role of RE and TM sublattices in the studied intermetallic compounds

Since the anisotropy of Y-containing compounds remains planar at all temperatures, by comparison of $\Delta \lambda$ curves for $Y_2Fe_{14-x}Co_xSi_2$ and $R_2Fe_{14-x}Co_xSi_2$ (R = Er, Tm) above $T > T_{SR}$ where planar anisotropy is dominated, we can deduce the contribution of rare-earth (RE) and transition-metal (TM) sublattices to the anisotropic magnetostriction of the later compounds. The results for compounds with x = 0 in Fig. 9(a) show that the value of $\Delta \lambda$ is larger in Tm than in Y-containing compounds up to T = 228 K and then the two curves crossover at 228 K. This behaviour implies that for $Tm_2Fe_{14}Si_2$ due to the influence of T_{SR} , Tm and TM have positive (increase) and then negative (decrease) magnetic moment coupling with temperature increase. This behaviour influences the magnetostriction parameters of the polycrystalline Tm-containing compound. Due to the non-magnetic Y-sublattice in Y-containing compounds the above behaviour does not occur and, therefore, the measured $\Delta \lambda$ is expected to be due to the effect of Fe-Fe sublattice interactions. For Er-containing compounds the behaviour may indicate that RE-TM coupling is increasing with temperature and decreasing beyond ~250K which can be attributed to the parastrictive temperature range. In contrast, as is clear from Fig. 9(b) for compounds with x = 4, the contribution of both RE and TM sublattices are added together. This is due to the fact that by substitution of Co atoms for Fe, the volume of the unit cell decreases. This in turn enhances the positive magnetostriction parameters of the RE sublattice.



Fig. 8. Temperature dependence of the volume magnetostriction of (a) $Tm_2Fe_{14}Si_2$ and (b) $Tm_2Fe_{10}Co_4Si_2$ compounds at selected magnetic fields.

It is to be noted that it is difficult to deduce the role of RE and TM sublattices below the spin reorientation transitions of these compounds by using the magnetostriction parameters of the polycrystalline Y-containing compound. To obtain accurate results it is necessary to investigate the magnetoelastic behaviour of $Y_2Fe_{14-x}Co_xSi_2$ single crystals.

4. Conclusion

We investigated the magnetoelastic properties of the offstoichiometric intermetallic compounds of $R_2Fe_{14-x}Co_xSi_2$ (R = Y, Er, Tm and x = 0, 4) with the R_2Fe_{17} -type structure using magnetostriction measurements. Influences of Si substitution on the studied compounds are as follow: (1) the anisotropic magnetostriction of $Y_2Fe_{14}Si_2$ shows sign change and reduction in its values compared to those for Y_2Fe_{17} below room temperature. Also, the value of $\Delta\lambda$ increases at least one order of magnitude at room temperature; (2) in $Er_2Fe_{14}Si_2$, anisotropic and volume magnetostrictions exhibit a sign change at spin reorientation temperature and (3) no sign change observed for $\Delta\lambda$ isothermal curves of $Tm_2Fe_{14}Si_2$ when passing through T_{SR} . Effects of Co substitution for Fe in $R_2Fe_{14}Si_2$ compounds show that: (1) anisotropic magnetostriction decreases in Y-containing compound; (2) the $\Delta\lambda$ curves of $Er_2Fe_{10}Co_4Si_2$ do not show any sign



Fig. 9. Comparison of temperature dependence of $\Delta\lambda$ for $\text{Er}_2\text{Fe}_{14-x}\text{Co}_x\text{Si}_2$ and $\text{Tm}_2\text{Fe}_{14-x}\text{Co}_x\text{Si}_2$ with $\text{Y}_2\text{Fe}_{14-x}\text{Co}_x\text{Si}_2$ compound (a) x = 0 and (b) x = 4, at highest applied field (1.5 T).

change around $T_{\rm SR}$ whereas the $\Delta V/V$ variations show opposite behaviour compared to that of ${\rm Er_2Fe_{14}Si_2}$ compounds and (3) the rare-earth sublattice contribution to the magnetostriction of ${\rm R_2Fe_{10}Co_4Si_2}$ is considerably larger than that for ${\rm R_2Fe_{14}Si_2}$ compounds.

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