



# Influence of low Co substitution on magnetoelastic properties of $\text{HoFe}_{11}\text{Ti}$ intermetallic compound

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## ABSTRACT

The thermal expansion and magnetostriction of  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  ( $x=0, 0.3, 0.7$  and  $1$ ) intermetallic compounds were measured, using the strain gauge method in the temperature range  $77\text{--}590\text{ K}$  under applied magnetic fields up to  $1.5\text{ T}$ . Results show that for samples with  $x=0$  and  $0.3$ , both linear thermal expansion and linear thermal expansion coefficient exhibit anomalies below the Curie temperature. Below room temperature, the spontaneous volume magnetostriction decreases with Co content. For all compounds studied, the anisotropic magnetostriction shows similar behaviour in the measured temperature range. The magnetostriction compensation occurs above room temperature in all samples. The volume magnetostriction shows a linear dependence on the applied field and by approaching the Curie temperature this trend changes to parastrictive behaviour. The results of the spontaneous magnetostriction are discussed based on the local magnetic moment model. The contribution of magnetostriction attributed to the magnetic sublattices R and T (Fe or Co) is discussed.

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## 1. Introduction

Intermetallic compounds based on rare-earth (R) and transition metal (T) elements are important from technological and fundamental point of view. Ternary intermetallic compounds of the type  $\text{RT}_{11}\text{M}$  ( $T=\text{Fe, Co or Mn}$  and  $M=\text{Ti, V, Cr, Mo, W, or Si}$ ) have been intensively investigated as the candidate for permanent magnet applications. All these compounds crystallise in the tetragonal  $\text{ThMn}_{12}$  structure with the space group  $I4/mmm$ , where the R atoms are located on one crystallographic site ( $2a$ ) and the T atoms occupy three non-equivalent sites ( $8f, 8i$  and  $8j$ ) [1–3]. Pseudo-ternary compounds of the type  $\text{RFe}_{11-x}\text{Co}_x\text{Ti}$  also crystallise in the  $\text{ThMn}_{12}$  structure [4]. In the past two decades, magnetic properties and magnetic structure of  $\text{RFe}_{11}\text{Ti}$  have been studied in order to investigate the nature of the field-induced magnetic phase transitions [5–7]. Many researches have also been performed on the investigation of magnetoelastic properties of intermetallic compounds based on  $\text{RFe}_{12-x}\text{M}_x$  [8–12]. In the previous study, we investigated the structural and magnetic properties of  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  compounds [13]. It was found that

lattice parameters decrease and Curie temperature ( $T_C$ ) increases with cobalt content. Spin reorientation transition as a result of the competition between anisotropies of Ho and T (Fe,Co) sublattices has been observed only for  $x=9$  and  $11$  compounds. However, the influence of Co concentration on the thermal expansion and magnetostriction of  $\text{RFe}_{11-x}\text{Co}_x\text{Ti}$  compounds has seldom been studied. To our knowledge, there is only one report on the thermal expansion of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  compounds that showed the variation of the thermal expansion anomaly in these compounds [14]. The influence of Co substitution on this anomaly in the presence of magnetic moment of a rare-earth element has not been investigated until now. In this work, we report the effect of low Co concentration on the magnetostriction and thermal expansion of  $\text{HoFe}_{11}\text{Ti}$  compound.

## 2. Experimental methods

The  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  ( $x=0, 0.3, 0.7$  and  $1$ ) polycrystalline samples were prepared by arc Ar atmosphere. Then, the buttons were re-melted in a high-frequency induction furnace equipped with a water-cooled copper crucible. To assure the homogeneity, the samples were subsequently wrapped into Ta foil, sealed into an evacuated quartz tube and annealed for 12 day at  $1050\text{ }^\circ\text{C}$  and quenched in air. X-ray diffraction (with  $\text{Cu-K}\alpha$  radiation) patterns

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of the annealed samples revealed the formation of single phase materials.

Thermal expansion and magnetostriction measurements were performed by using a strain gauge method. The gauges were placed on disc-shaped samples (10 mm diameter, 3 mm thick), which were cut from the annealed bulk ingots. Magnetic fields were applied up to 1.5 T, with temperature ranging from 77 to 590 K. The thermal expansions  $\Delta l/l = [l(T) - l(77 \text{ K})]/l(77 \text{ K})$  were deduced by measuring the relative change in length of the samples versus temperature. The magnetostriction was measured (with an accuracy of  $2 \times 10^{-6}$ ) parallel (longitudinal magnetostriction,  $\lambda_l$ ) and perpendicular (transverse magnetostriction  $\lambda_t$ ) to the field direction, thus allowing to deduce the anisotropic ( $\Delta\lambda = \lambda_l - \lambda_t$ ) and the volume ( $\Delta V/V = \lambda_l + 2\lambda_t$ ) magnetostriction. It should be emphasised that no significant difference was observed between the strains measured in the plane and perpendicular to the plane of the disc of the samples, suggesting the absence of any preferred orientation effects.

### 3. Results and discussion

The structural and magnetic properties of these samples were investigated by authors in Ref. [13]. It was shown that the samples are single phase [13].

The linear thermal expansion curves,  $\Delta l/l(T)$ , of  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  compounds are shown in Fig. 1. The Curie temperatures of the

samples with  $x=0$  and 0.3 are indicated by arrows. It can be seen that, for  $x=0$ , both  $\Delta l/l(T)$  and linear thermal expansion coefficient,  $\alpha(T) = (d/dT)[\Delta l/l(T)]$ , exhibit an anomalous behaviour and Invar effect near  $T_C$ . Beyond  $T_C$  in the paramagnetic region,  $\Delta l/l(T)$  behaves linearly. Sample with  $x=0.3$  shows similar behaviour, but without Invar effect. For samples with  $x=0.7$  and 1, the Curie temperature (determined by magnetisation method) is at 593 K and 623 K, respectively [13]; which are higher than the maximum temperature allowed to apply to the strain gauge. Therefore, the curves for these samples show only the behaviour in the magnetic ordering state. The spontaneous linear magnetostriction,  $[\Delta l/l(T)]_m$ , was determined by subtracting the lattice thermal expansion,  $[\Delta l/l(T)]_{\text{latt}}$ , from the total experimental linear thermal expansion,  $[\Delta l/l(T)]_{\text{exp}}$ , i.e.  $[\Delta l/l(T)]_m = [\Delta l/l(T)]_{\text{exp}} - [\Delta l/l(T)]_{\text{latt}}$ . The lattice contribution (the dashed lines in Fig. 1) is obtained by extrapolating the paramagnetic range according to the Debye theory with the Debye temperature,  $T_D = 450 \text{ K}$  in this case [8–10], and the Grüneisen relation,  $\alpha(T) = \kappa\gamma C_V/3$ , where  $\alpha$  is the phonon anharmonic thermal expansion coefficient,  $\kappa$  is the isothermal compressibility,  $\gamma$  is the Grüneisen parameter, and  $C_V$  is the specific heat. With assumption that  $\Delta l/l(T)$  is isotropic, spontaneous volume magnetostriction will be  $\omega_s(T) = 3[\Delta l/l(T)]_m$ , which is defined as the relative volume difference between the magnetic and paramagnetic states. Fig. 2 shows the temperature dependence of  $\omega_s(T)$  for  $\text{HoFe}_{11}\text{Ti}$  compound which illustrates decrease of  $\omega_s$  from  $9.0 \times 10^{-3}$  to about 0 at  $T_C$ . The nonzero remaining values of  $\omega_s$  above  $T_C$  reflect the existence of a short-range magnetic ordering in the paramagnetic state, similar to some other 1–12 compounds [8–10]. Comparison of  $\omega_s$  in polycrystalline sample of  $\text{HoFe}_{11}\text{Ti}$  with that of  $\text{YFe}_{11}\text{Ti}$  in Ref. [14] shows that  $\omega_s$  is smaller in the Ho containing compound. This indicates that the volume dependence of magnetisation ( $dM/d\omega$ ) (volume dependence of the magnetic moments) in  $\text{HoFe}_{11}\text{Ti}$  is lower than that of  $\text{YFe}_{11}\text{Ti}$  compound. A similar behaviour was also determined for the pressure effect on the magnetisation of these compounds which shows that  $dM/dP$  is equal to  $-0.065$  and  $-0.056$  ( $\mu_B/f.u.$ )  $\text{kbar}^{-1}$  for Y- and Ho-containing compounds, respectively [15]. Therefore, these results may indicate that Ho sublattice moment induces a negative effect on spontaneous volume magnetostriction. Fig. 2 shows the temperature dependence of  $\omega_s(T)$  for sample with  $x=0.3$ . As temperature decreases below  $T_C$ ,  $\omega_s$  increases up to 170 K and then decreases gradually. The presence of the maximum in  $\omega_s$  around 170 K can be

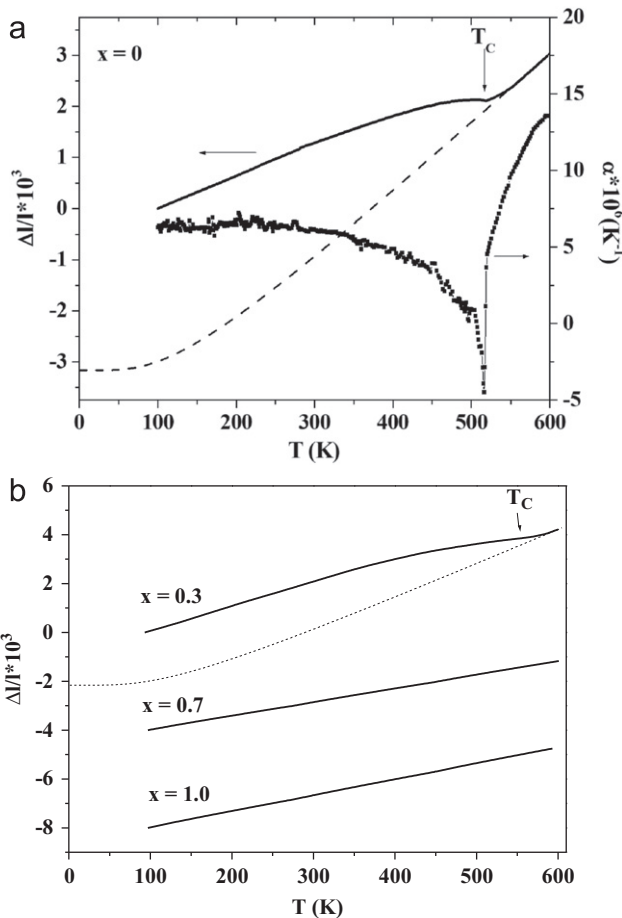


Fig. 1. (a) Linear thermal expansion  $\Delta l/l(T)$  and linear thermal expansion coefficient  $\alpha(T)$  for  $\text{HoFe}_{11}\text{Ti}$  compound, (b)  $\Delta l/l(T)$  for  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  ( $x=0.3, 0.7$  and  $1$ ) compounds. The dashed lines represent the calculated phonon contribution to the thermal expansion as discussed in the text. The arrows indicate the Curie temperatures.

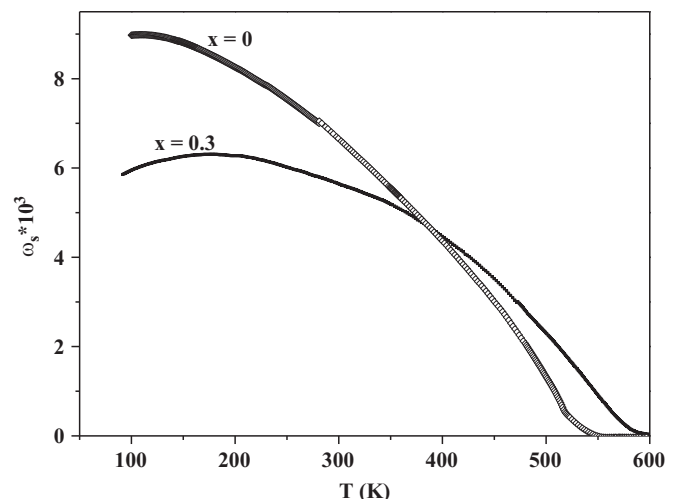


Fig. 2. Temperature dependence of the spontaneous volume magnetostriction ( $\omega_s$ ) for  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  ( $x=0$  and  $0.3$ ) compounds. The arrows indicate the Curie temperatures.

attributed to the linear and non-Invar behaviour of the thermal expansion in low temperatures. Such behaviour is also observed in  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  compounds [14]. Finally, as shown in Fig. 2,  $\omega_s$  decreases with Co content in  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  compounds.

In order to investigate the effect of Co substitution on  $\omega_s(T)$ , we use the local moment model (the Heisenberg model). In this model, the volume difference ( $\omega_s$ ) due to the magnetic coupling is given by the two isotropic spin correlation functions  $\langle \mathbf{m}_i \cdot \mathbf{m}_j \rangle$  [16]

$$\omega_s = \kappa \sum_{i,j} C^{\text{int}} \langle \mathbf{m}_i \cdot \mathbf{m}_j \rangle \quad (1)$$

where  $\kappa$  is the compressibility,  $C^{\text{int}}$  is the magnetovolume coupling constant due to the volume dependence of exchange interaction and  $i, j$  are the lattice sites. It is well known that in R–T compounds, there are three exchange interactions included  $J_{\text{TT}}$  between the transition metal spins,  $J_{\text{RT}}$  between the rare earth spin and transition metal spins, and  $J_{\text{RR}}$  between rare earth spins. Since the Curie temperature is nearly equal in  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  and  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  compounds; therefore,  $J_{\text{TT}}$  is dominant in comparison to  $J_{\text{RT}}$ , whereas  $J_{\text{RR}}$  contribution can be neglected [13,14]. Previous results showed that in  $\text{RFe}_{12-x}\text{M}_x$  alloys,  $\text{M}(\text{=Ti, Mo, etc.})$  atoms substitute preferentially into the  $8i$  site. This site is corresponding to  $6c$  (dumbbell) site in  $\text{Th}_2\text{Zn}_{17}$ -type [17]. The critical bond length between a positive exchange interaction (ferromagnetic) and a negative exchange interaction (antiferromagnetic) is about  $2.45 \text{ \AA}$ . In  $\text{ThMn}_{12}$  structure, two bond lengths of  $8i$ – $8i$  (in the  $ab$ -plane) and  $8f$ – $8f$  (half of the lattice parameter  $c$  and along the  $c$ -axis) are smaller than  $2.45 \text{ \AA}$  (critical bond length). Therefore, the coupling between these sites is strong with negative value and responsible for the spontaneous volume magnetostriction.

Our previous results show that the introduction of Co into the  $\text{HoFe}_{11}\text{Ti}$  alloy resulted in a decrease in lattice parameters [13]. Consequently, it can be expected that the bond lengths of  $8i$ – $8i$  and  $8f$ – $8f$  decrease and negative exchange interaction increases. However, the total  $J_{\text{TT}}$  is not only determined by the exchange parameter  $J_{\text{TT}}^{ij}$  (exchange integral between  $\text{T}\{i\}$  and  $\text{T}\{j\}$ , where  $\{i\}$  and  $\{j\}$  are  $\text{T}(\text{Fe, Co})$  atoms for equivalent and non-equivalent sites) but also by  $\text{T}$  magnetic moment and the number of  $\text{T}$  nearest neighbours. Indeed, based on the Wigner–Seitz cell calculation, Co atoms occupy  $8f$  sites [13]. Thus, although the exchange parameter  $J_{\text{FeFe}}^{ij}$  increases by lattice contraction, the magnetic moment and the number of  $\text{Fe}$ – $\text{Fe}$  pairs in  $8f$  site decreases by Co substitution. The contribution of  $\text{Co}$ – $\text{Co}$  interaction to  $J_{\text{TT}}$  is positive and independent of the interatomic distances. This explanation is consequence of a little change of  $T_C$  in  $\text{Co}$ -rich  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  compounds [13]. Finally, the decrease in magnetovolume effect in  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  compounds with Co substitution indicates that the magnetovolume coupling constant ( $C^{\text{int}}$ ) decreases, i.e. the total exchange interaction is independent of the interatomic distance. A similar trend has been observed in  $\text{R}_2\text{Fe}_{14-x}\text{Co}_x\text{B}$  ( $\text{R}=\text{Y}$  and  $\text{Er}$ ) compounds [18,19]. Moreover, from the decrease of  $\omega_s$  value, it can be concluded that by increasing Co content the compounds tend to become strong ferromagnets. This conclusion can be understood by following the magnetic valence model and the theoretical calculations of the density of states for  $\text{Fe}$  and  $\text{Co}$  [20,21].

Lee and Pourarian employed the local moment model, using the spontaneous volume magnetostriction in a two sublattice approximation described in the case of R–T intermetallic compounds as [22]

$$\omega_s = n_{\text{TT}}\mu_{\text{T}}^2 + n_{\text{RT}}\mu_{\text{R}}\mu_{\text{T}} + n_{\text{RR}}\mu_{\text{R}}^2 \quad (2)$$

where  $\mu_{\text{T}}$  and  $\mu_{\text{R}}$  are the average magnetic moments of the  $\text{T}$  and  $\text{R}$  atoms,  $n_{\text{TT}}$  and  $n_{\text{RR}}$  are the magnetoelastic coupling coefficients

in the  $\text{T}$  and  $\text{R}$  sublattices, respectively, and  $n_{\text{RT}}$  is the intersublattice coupling coefficient between  $\text{R}$  and  $\text{T}$  atoms. For intermetallic compounds with high content of  $3d$  metals, it is possible to neglect the last term in relation (2) [10]. Using relation (2) and the values of  $\mu_{\text{Ho}}, \mu_{\text{Fe}}$  and  $n_{\text{FeFe}}$  at  $5 \text{ K}$  from [13,14], the intermetallic coupling coefficient,  $n_{\text{HoFe}}$ , is obtained as  $-1.7 \times 10^{-5} \mu_{\text{B}}^{-2}$  at  $5 \text{ K}$  that is insignificant in comparison to  $n_{\text{FeFe}}$ . In this calculation, we can use the linear fit of thermal expansion experimental curve at low temperature for obtaining  $\omega_s$  at  $5 \text{ K}$ .

Fig. 3 shows the anisotropic magnetostriction ( $\Delta\lambda$ ) of  $\text{HoFe}_{11}\text{Ti}$  compound as a function of applied field at some selected temperatures. It is clear that below room temperature  $\Delta\lambda$  monotonically increases with negative sign under applied field. In this compound, the anisotropy field is larger than  $1.5 \text{ T}$  below room temperature (RT). Thus, the tendency to saturation is not observed in this temperature range. Saturation behaviour starts approximately from  $T=330 \text{ K}$ . This behaviour may be attributed to the reduction of the magnetocrystalline anisotropy with increasing temperature.

The temperature dependence of  $\Delta\lambda$  for  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  ( $x=0, 0.7$  and  $1$ ) compounds at applied field of  $1.5 \text{ T}$  is shown in Fig. 4.

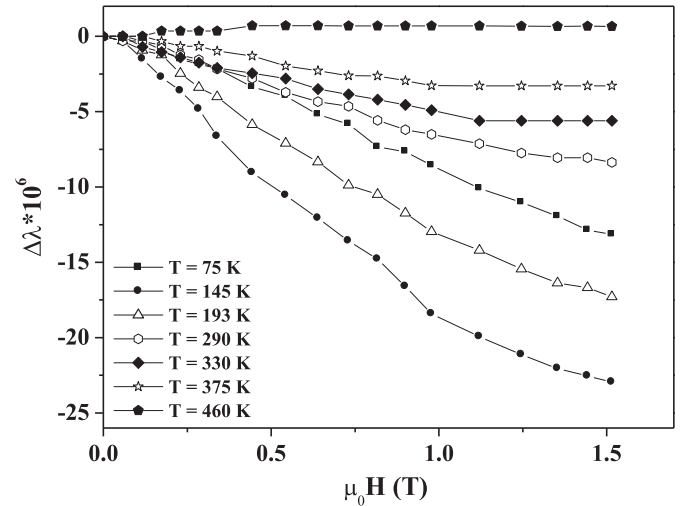


Fig. 3. Field dependence of the anisotropic magnetostriction of  $\text{HoFe}_{11}\text{Ti}$  compound at different temperatures. In this and all the following figures, the lines are guides for the eyes.

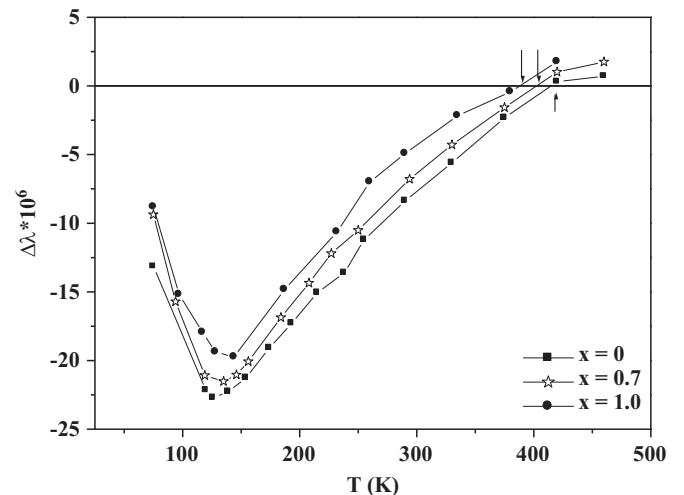


Fig. 4. Temperature dependence of the anisotropic magnetostriction of  $\text{HoFe}_{11-x}\text{Co}_x\text{Ti}$  ( $x=0, 0.7$  and  $1$ ) compounds at the applied magnetic field of  $1.5 \text{ T}$ . The arrows refer to the compensation point ( $T_0$ ).

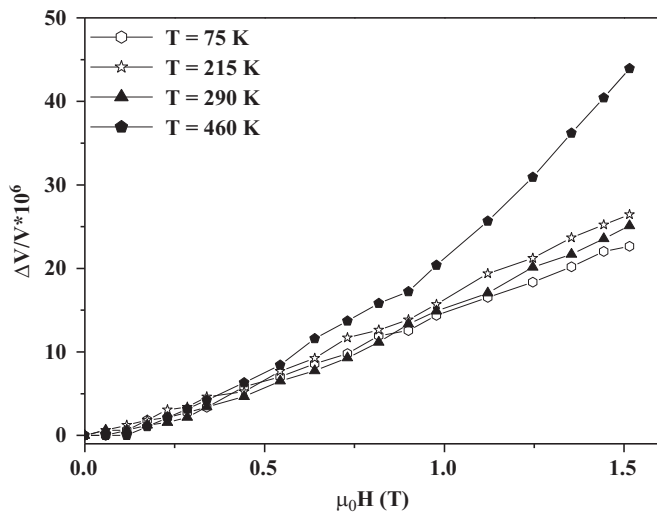


Fig. 5. Field dependences of the volume magnetostriction of HoFe<sub>11</sub>Ti compound at some typical temperatures.

All compounds show similar behaviour in the whole temperature range, but the absolute values of  $\Delta\lambda$  decrease slightly with Co content. As one can observe the anisotropic magnetostriction has a minimum around  $T=125$  K and then increases. Finally, magnetostriction compensation occurs at a point ( $T_0$ ) above room temperature. It is seen that  $T_0$  decreases with Co substitution. Previous results on magnetostriction of RFe<sub>11</sub>Ti (Y, Tb and Dy) compounds show that Fe sublattice contribution to magnetostriction is positive in these compounds [11,12]. A similar trend was also reported for Fe sublattice contribution to the magnetostriction of YFe<sub>12-x</sub>V<sub>x</sub> compounds [23]. Therefore, using our results on  $\Delta\lambda$  of HoFe<sub>11</sub>Ti, one can deduce that Ho sublattice contribution to magnetostriction of this compound is negative. Hence, observed minimum in  $\Delta\lambda$  curve could be attributed to the gradually increasing (decreasing) contribution of Fe (Ho) sublattice to  $\Delta\lambda$ . Such behaviour for Fe sublattice is observed in YFe<sub>10</sub>V<sub>2</sub> compound as well [24]. The observed positive value of  $\Delta\lambda$  in these compounds beyond compensation point ( $T_0$ ) can be attributed to the natural reduction of the anisotropy of Ho sublattice and is due to Fe-sublattice anisotropy. Moreover, the decrease of  $T_0$  by Co substitution in HoFe<sub>11-x</sub>Co<sub>x</sub>Ti compounds shows that Co induces positive anisotropic magnetostriction in these compounds and enhances contribution of T-sublattice to  $\Delta\lambda$ .

The volume magnetostriction ( $\Delta V/V$ ) of the HoFe<sub>11</sub>Ti compound is plotted versus applied field at some selected temperatures shown in Fig. 5. Except at high temperatures, the  $\Delta V/V$  curves of all compounds show almost linear dependence with field.  $\Delta V/V$  also increases monotonically with field. Moreover, as expected for the parastrictive behaviour, for temperatures near  $T_C$  the  $\Delta V/V$  curves tend to have a quadratic dependence on the applied field [25]. This behaviour occurs at higher temperatures for HoFe<sub>11-x</sub>Co<sub>x</sub>Ti compounds. On the other hand,  $\Delta V/V$  decreases with increasing Co concentration in these compounds. The results of  $\omega_s$  indicate that the volume dependence of the magnetic interaction weakens with Co substitution. A similar trend has also been observed in Er<sub>2</sub>Fe<sub>14-x</sub>Co<sub>x</sub>B and Tm<sub>2</sub>Fe<sub>14-x</sub>Co<sub>x</sub>Si<sub>2</sub> compounds [18,26].

#### 4. Conclusion

The influence of Co substitution on the magnetoelastic properties of HoFe<sub>11</sub>Ti polycrystalline compound has been investigated. The spontaneous volume magnetostriction and Invar effect

decrease with Co substitution. Results show that Ho sublattice has negative contribution to spontaneous volume magnetostriction in this compound. Magnetostriction compensation occurs above room temperature in HoFe<sub>11-x</sub>Co<sub>x</sub>Ti compounds. The absolute values of  $\Delta\lambda$  decrease slightly with Co content. From our results on  $\Delta\lambda$  of HoFe<sub>11</sub>Ti, it can be deduced that Ho sublattice contribution to magnetostriction is negative and the observed positive value of  $\Delta\lambda$  in HoFe<sub>11-x</sub>Co<sub>x</sub>Ti compounds beyond the compensation point is due to the T-sublattice anisotropy. The volume magnetostriction decreases with increasing Co concentration in these compounds.

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