



# A study on magnetoelastic properties of $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$ ( $x=0, 3, 6$ ) compounds

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## ABSTRACT

In this work, The magnetoelastic properties of polycrystalline samples of  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) intermetallic compounds are investigated by means of linear thermal expansion and magnetostriction measurements in the temperature range of 77–515 K under applied magnetic fields up to 1.5 T. The linear thermal expansion increases with the Co content. The well-defined anomalies observed in the linear thermal expansion coefficients for  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) compounds are associated with the magnetic ordering temperature for  $x=0$  and spin reorientation temperatures for  $x=3, 6$ . Below transition temperatures, the value of the longitudinal magnetostriction ( $\lambda_{\text{Pa}}$ ) at 1.6 T increases with Co content.

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## 1. Introduction

Intermetallic compounds  $\text{R}_3(\text{Fe,M})_{29}$  (3:29) crystallize in monoclinic system with  $A2/m$  space group [1]. There are two formula units and 64 atoms per unit cell with the rare earth ions occupy two nonequivalent crystallographic sites (2a and 4i), and the Fe atoms occupy 11 sites (one 2c, one 4e, one 4g, four 4i and four 8j) [2].

Since Co has different electronic structure than Fe, and therefore a different local anisotropy, the structural and magnetic properties of 3:29 compounds are strongly affected by the partial substitution of Fe by Co atoms. In replacing Fe by small amount of Co [3] (Ni [4]) atoms, they substitutes preferentially at  $\text{Fe}_1$  (2c),  $\text{Fe}_8$  (8j) and  $\text{Fe}_{11}$  (4e) sites, where the Fe atoms couple with a negative exchange interaction with their neighbor atoms. The substitution of Co increases net exchange by reducing the negative contribution of antiferromagnetically coupled sites on the 3d sublattice and thereby  $T_C$  increases rapidly [3,5]. The Ti, Mo and V atoms substitute in  $\text{Fe}_2$  (4i),  $\text{Fe}_3$  (4i) and  $\text{Fe}_6$  (4g) sites where the dumb-bell Fe atoms are arranged in a direction close to the c-axis, while Co atoms strongly avoid these sites [3,6]. However, for large concentrations of Co atoms they occupy the dumb-bell positions [7].

In  $\text{Y}_3(\text{Fe}_{1-x}\text{Co}_x)_{27.5}\text{V}_{1.5}$  compounds, the EMD for  $x=0$  is of easy plane type [8], but for  $x=0.1$ –0.4, as the cobalt content increases the easy magnetization direction (EMD) gradually turns without bending to the outside of the basal plane [9]. The EMD determined experimentally for  $\text{Tb}_3(\text{Fe,V})_{29}$  [10] compounds at room temperature is along the *b*-axis ([0 1 0] direction) of the monoclinic structure. In  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  compounds EMD gradually turns in the basal plane for  $x=3$  but for  $x=6$  bend to the outside of the basal plane and a tilted magnetic structures are formed [11]. In addition, the magnetocrystalline anisotropy measurements of  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  compounds using the singular point detection (SPD) technique indicate this change of the magnetic anisotropy due to the increase of the Co content [12].

A spin reorientation transition at 160 K is reported for  $\text{Tb}_3(\text{Fe}_{1-x}\text{V}_x)_{29}$  polycrystalline compound [13]. However, this result was failed by magnetization measurements along the hard axis of a  $\text{Tb}_3(\text{Fe}_{1-x}\text{V}_x)_{29}$  single crystal [12]. A uniaxial anisotropy favorable for permanent magnet applications is expected to occur in the 3:29 system in [2 0 –1], [1 0 2] and [0 1 0] directions [13]. The EMD determined experimentally for  $\text{Tb}_3(\text{Fe,V})_{29}$  compound at room temperature is [0 1 0] direction [10]. Since the anisotropy of the 3d sublattice is planar ([2 0 –1]) [13], therefore the 4f anisotropy overcomes the 3d anisotropy at room temperature. In  $\text{Tb}_3(\text{Fe}_{1-x}\text{Co}_x)_{27.4}\text{V}_{1.6}$  ( $x=0.0$ –0.4) series the spin reorientation phenomena is observed only in  $x=0.1$ –0.3 which by increasing Co content the transition temperature decreases from 473 K for  $x=0.1$  to 393 K for  $x=0.3$ , and disappears in the case of  $x=0.4$  [10]. Gholizadeh et al. [11], observed the type 2 FOMP for  $\text{Tb}_3$

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(Fe<sub>28-x</sub>Co<sub>x</sub>) V<sub>1.0</sub> ( $x=0, 3, 6$ ) polycrystalline compounds using the singular point detection (SPD) technique from 5 to 300 K. This transition is believed to be attributed to an occurrence of canting angles between the magnetic sublattices during the magnetization process.

Thermal expansion measurements on R<sub>3</sub> (Fe,Ti)<sub>29</sub> (R=Ce, Nd,Gd and Tb) [13–18], Ce<sub>3</sub> (Fe,Cr)<sub>29</sub> [19], and Gd<sub>3</sub> (Fe,Co,Cr)<sub>29</sub> compounds [20], and magnetostriction measurements on Y<sub>3</sub>Fe<sub>27.2</sub>Cr<sub>1.8</sub> and Ce<sub>3</sub>Fe<sub>25</sub>Cr<sub>4</sub> compounds [21] have been previously reported. In R<sub>3</sub> (Fe,M)<sub>29</sub> compounds, invar-type behavior was reported with M=Ti [14]. The stabilizing element (M=Cr,Ti, V...) plays a crucial role in the thermal expansion behavior of 3:29 intermetallic system, as in the case of some RFe<sub>12-x</sub>M<sub>x</sub> compounds [22]. The low symmetry of the R<sub>3</sub> (Fe<sub>1-x</sub>M<sub>x</sub>)<sub>29</sub> structure implies that there are one easy and two hard directions. Anisotropy field is the magnetic field needed to rotate the magnetization vectors from an easy to a hard direction. There are two anisotropy fields  $H_a$  and  $H_a'$  ( $H_a > H_a'$ ) in R<sub>3</sub> (Fe,M)<sub>29</sub> compounds. The magnetostriction behavior of Y<sub>3</sub>Fe<sub>27.2</sub>Cr<sub>1.8</sub> and Ce<sub>3</sub>Fe<sub>25</sub>Cr<sub>4</sub> compounds exhibits a minimum at different applied fields that have been attributed to  $H_a'$  anisotropy fields [21].

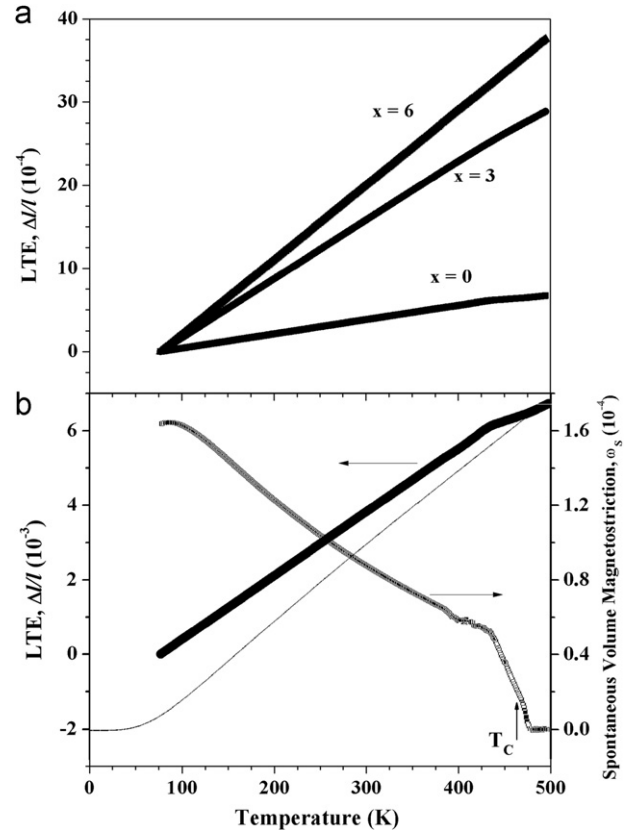
Previously, we have reported the measurements of anisotropy field ( $H_a$ ) as well as critical field ( $H_{cr}$ ) of Tb<sub>3</sub> (Fe<sub>28-x</sub>Co<sub>x</sub>) V<sub>1.0</sub> ( $x=0, 3, 6$ ) compounds using the singular point detection (SPD) technique from 5 to 300 K [11]. In this work, the effect of Co substitution on the magnetoelastic properties of Tb<sub>3</sub> (Fe<sub>28-x</sub>Co<sub>x</sub>) V<sub>1.0</sub> ( $x=0, 3, 6$ ) compounds between 77 and 515 K are studied via magnetostriction and thermal expansion measurement using strain gauge method which so far has not been investigated.

## 2. Experimental

Tb<sub>3</sub> (Fe<sub>28-x</sub>Co<sub>x</sub>) V<sub>1.0</sub> ( $x=0, 3, 6$ ) compounds were prepared by arc melting of high-purity (at least 99.9%) of the constituent elements in a water-cooled copper boat. In order to maximize the amount of 3:29 phase, the ingots were subsequently annealed in sealed quartz tubes under a protective argon atmosphere at 1323 K for a period of one week and then water-quenched to room temperature. The structural characterization of compounds by X-ray powder diffraction is evidence for a monoclinic Nd<sub>3</sub> (Fe,Ti)<sub>29</sub>-type structure (A2/m space group) [11]. Magnetostriction and linear thermal expansion (LTE) measurements were carried out by the standard strain gauge method on disk-shaped samples of 6 mm diameter and 3 mm thickness in magnetic fields up to 1.5 T, and temperatures ranging from 77 to 515 K. The spontaneous volume magnetostriction  $\omega_s$  was determined for  $\omega_s = 3[(\Delta l/l)_{\text{exp}} - (\Delta l/l)_{\text{lat}}]$ , where  $(\Delta l/l)_{\text{exp}} = ((l(T) - l(77 \text{ K})) / l(77 \text{ K}))$  and  $(\Delta l/l)_{\text{lat}}$  is the lattice contribution that can be obtained from the extrapolation of the paramagnetic regime of the LTE curve, where only the non-magnetic inharmonic phonon contribution is expected. The extrapolation has been performed using the Grüneisen–Debye model, with a Debye temperature  $T_D = 450 \text{ K}$ . Magnetostrictions were measured (with an accuracy of  $2 \times 10^{-6}$ ) parallel (longitudinal magnetostriction,  $\lambda_l$ ) and normal (transverse magnetostriction,  $\lambda_t$ ) to the applied field direction. It should be emphasized 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

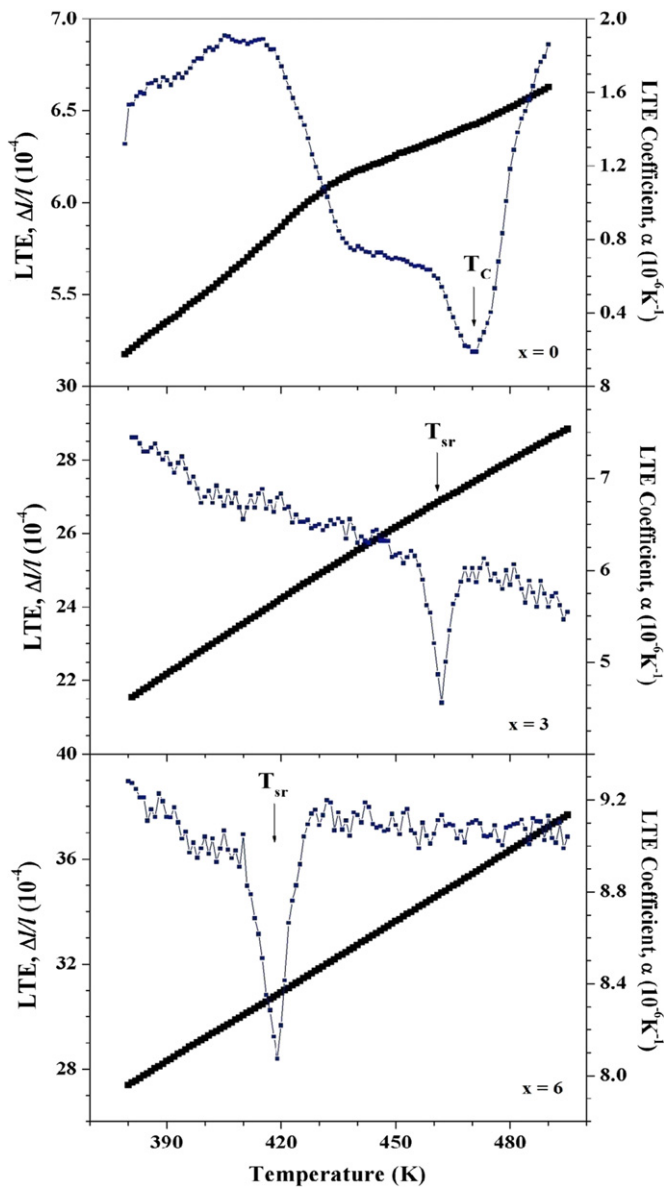
In Fig. 1(a), the measured linear thermal expansion (LTE) for Tb<sub>3</sub> (Fe<sub>28-x</sub>Co<sub>x</sub>) V<sub>1.0</sub> ( $x=0, 3, 6$ ) compounds and Fig. 1(b) temperature dependence of the linear thermal expansion (LTE) and spontaneous volume magnetostriction ( $\omega_s$ ) of Tb<sub>3</sub>Fe<sub>28</sub>V<sub>1.0</sub> are



**Fig. 1.** (a) The measured linear thermal expansion (LTE) for Tb<sub>3</sub> (Fe<sub>28-x</sub>Co<sub>x</sub>) V<sub>1.0</sub> ( $x=0, 3, 6$ ) compounds. (b) Temperature dependence of the linear thermal expansion and spontaneous volume magnetostriction of Tb<sub>3</sub>Fe<sub>28</sub>V<sub>1.0</sub>. The thin line represents the lattice LTE.

shown from 77 to 500 K. It is clear that LTE increases with the Co content. The temperature dependence of LTE is a consequence of superposition of contributions of phonons and magnons. In Fig. 1(b), an additional contribution to LTE (represented by a thin line) of Tb<sub>3</sub>Fe<sub>28</sub>V<sub>1.0</sub> is observed. This latter contribution is resulting from  $\omega_s$ . The value of  $\omega_s$  decreases from  $1.65 \times 10^{-3}$  to near zero as temperature increases from 77 K to  $T_C$ . The difference in the average values of magnetic moments of 3d sublattice in R<sub>3</sub> (Fe,Co,M)<sub>29</sub> compounds has an influence on the different values of  $\omega_s$ . Wang et al. [20] reported that in Gd<sub>3</sub> (Fe,Co,Cr)<sub>29</sub> compounds, the value of  $\omega_s$  decreases with increasing Co content and the 3d magnetic moment. Since in R<sub>3</sub> (Fe,Co,M)<sub>29</sub> compounds, the largest magnetovolume effect is observed in the Fe rich compounds, therefore our results indicate that the phonon contribution increases with the Co content. It can be noted that the value of unit cell volume of these compounds has influence on the different values and signs of the Fe–Fe exchange interactions.

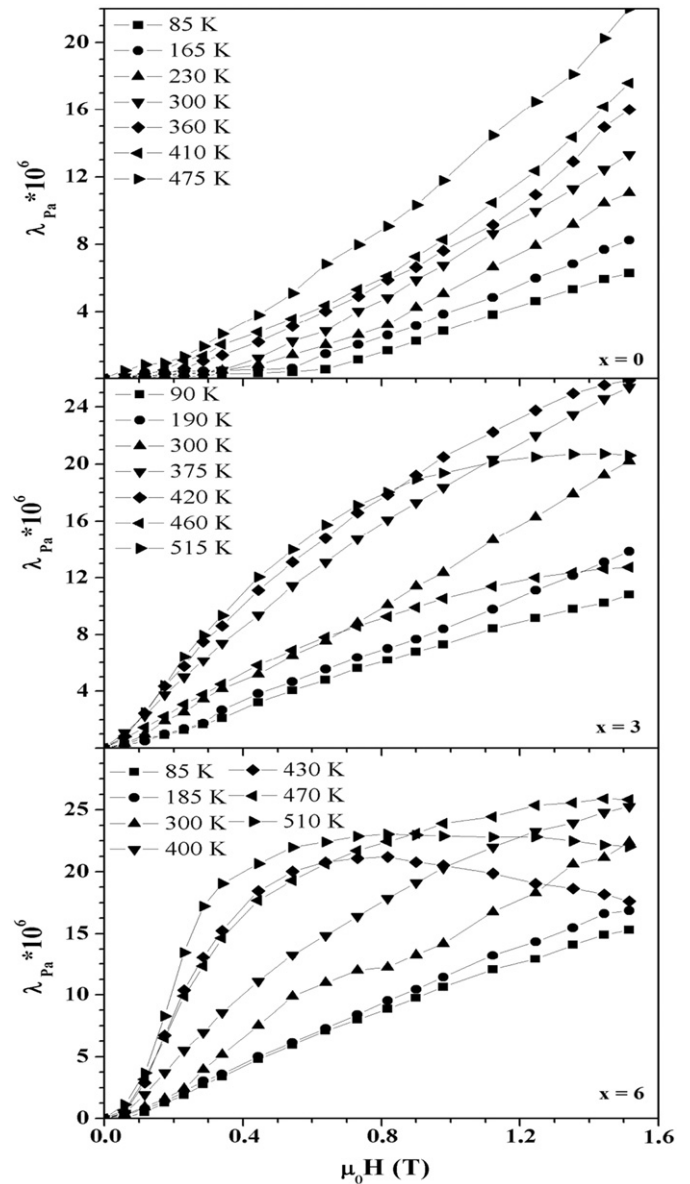
LTE of the studied compounds from 380 to 500 K, and their calculated linear thermal expansion coefficient,  $\alpha(T)$ , are displayed in Fig. 2. In addition, an anomalous invar-type behavior observe near  $T_C$  in  $\alpha(T)$  for Tb<sub>3</sub>Fe<sub>28</sub>V<sub>1.0</sub>. The average  $\alpha(T)$  below the magnetic phase transitions increase with increasing Co content. The well-defined anomalies observed at 468, 471 and 418 K for  $x=0, 3, 6$ , respectively, are associated with the magnetic ordering temperature for  $x=0$  and spin reorientation temperatures ( $T_{sr}$ ) for  $x=3, 6$ . The occurrence of spin reorientation transition originates from competition between the magnetocrystalline anisotropy of the 3d and 4f sublattices. In addition, the decrease in ( $T_{sr}$ ) can be attributed to a weakening of the rare-earth sublattice magneto crystalline anisotropy with the increase of Co content.



**Fig. 2.** The measured linear thermal expansion (LTE) and the LTE coefficient,  $\alpha$ , for  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) compounds presented from 380 to 500 K.

For all of compounds, isothermal curves of the longitudinal magnetostriction,  $\lambda_{\text{Pa}}$ , versus magnetic field at selected temperatures are shown in Fig. 3.  $\lambda_{\text{Pa}}$  shows a unique parabolic behavior at all temperatures. Also, the sign of curvature of isothermal curves that is positive for the  $x=0$ , changes to negative in the  $x=3, 6$  (Fig. 3). As shown in Fig. 3, below  $T_{\text{sr}}$ , the value of  $\lambda_{\text{Pa}}$  at maximum applied field increases with the Co content. However, the external field of 1.5 T is not sufficient to reach magnetic saturation. Therefore, for these samples magnetostriction measurements at higher external fields will be necessary. A clear change occurs around  $T_{\text{sr}}$  in the curves of the samples  $x=3, 6$ . At above this temperature, the saturation for  $x=6$  sample begins at lower field than for  $x=3$ .

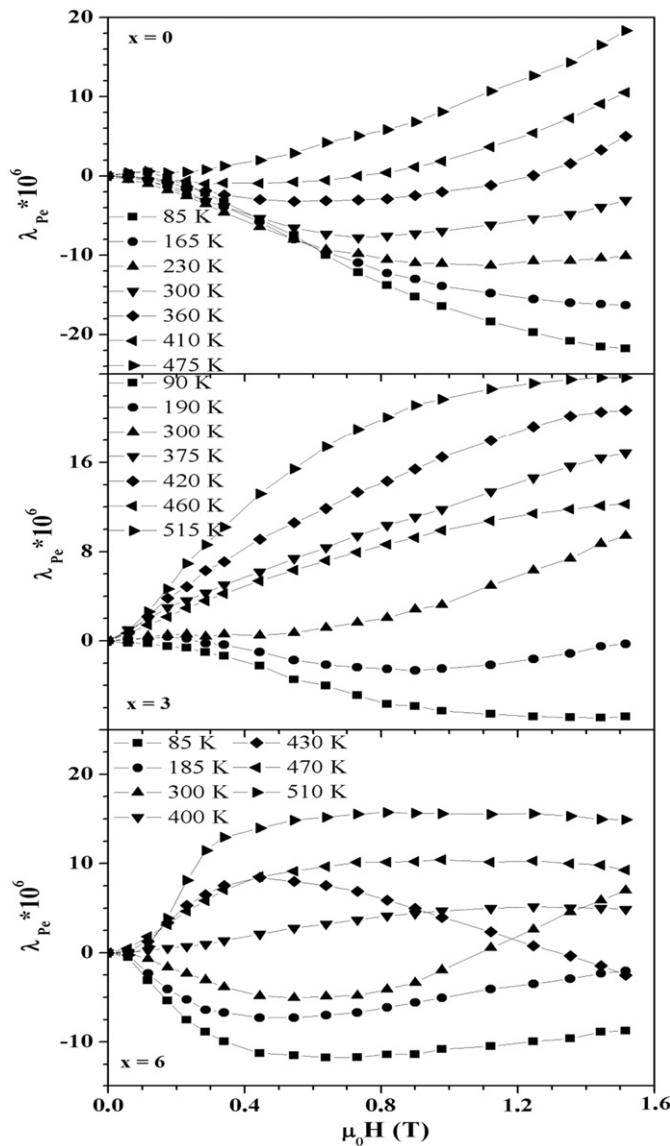
In Fig. 4 the transverse magnetostriction,  $\lambda_{\text{Pe}}$ , for  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) compounds are plotted versus magnetic field at selected temperatures.  $\lambda_{\text{Pe}}$  shows parabolic behavior at all temperatures. In addition, the sign of curvature of isothermal curves that is positive for all of the samples, changes to negative for  $x=3, 6$  from near  $T_{\text{sr}}$  to above it (Fig. 4).  $\lambda_{\text{Pe}}$  exhibits a minimum at different applied fields. However, it increases linearly in higher applied field. Concerning magnetization processes, one observes a



**Fig. 3.** Isothermal curves of the longitudinal magnetostriction of  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) compounds as a function of applied field at selected temperatures.

linear correspondence between the magnetostrictive effects and the rotation of the easy magnetization axis against the magnetic anisotropy forces.

The low symmetry of the  $\text{R}_3(\text{Fe}_{1-x}\text{M}_x)_{29}$  structure implies that there are one easy and two hard directions. Anisotropy field is the magnetic field needed to rotate the magnetization vector from an easy to a hard direction. For  $\text{Tb}_3(\text{Fe},\text{V})_{29}$ , the EMD is parallel to the  $b$ -axis ( $[0\ 1\ 0]$  direction of monoclinic structure), and the hard directions are along the  $c$  ( $[2\ 0\ -1]$ ) and  $a$  ( $[1\ 0\ 2]$ ) directions. The latter is the hardest magnetization direction of the compound at all the investigated temperatures [10,12]. In addition, the anisotropy field along the  $a$ -axis ( $H_a^a$ ) is larger than that of the  $c$ -axis ( $H_a^c$ ) [23]. In  $\text{R}_3(\text{Fe},\text{M})_{29}$  compounds with  $\text{M}=\text{V}$ ,  $H_a^c$  was not observed by SPD technique because of the occurrence of multifold twinning in the crystal [11,24]. However, assuming that there is one EMD along the  $b$ -axis and hard magnetization direction (HMD) in  $b$ - $c$  plane and one HMD along  $a$ -axis in our monoclinic compounds, we can conclude that at low temperatures, where the applied field ( $H$ ) is smaller than  $H_a^c$  the magnetization vector rotates in the  $b$ - $c$  plane, and  $\lambda_{\text{Pe}}$  has negative values and increases



**Fig. 4.** Temperature dependence of the transverse magnetostriction of  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) compounds as a function of applied field at selected temperatures.

with the applied field. When  $H > H_a^c$  the magnetic moments rotate toward the  $a$ -axis and  $\lambda_{pe}$  tends to change towards the positive values. Finally, the magnetization vector rotates toward the applied field resulting in a forced magnetostriction. In addition, the decrease in the minimum of  $\lambda_{pe}$  with the increase of Co content and temperature in  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  compounds may be attributed to decreasing of  $H_a^c$  magnetic anisotropy.

#### 4. Summary

Magnetostriction and linear thermal expansion of  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) compounds were reported. The measurements show that:

- (i) Lattice thermal expansion and consequently LTE increase with the Co content.
- (ii) The well-defined anomalies observed in LTE coefficients at 468, 471 and 418 K for  $x=0, 3, 6$ , respectively, are associated with the magnetic ordering temperature for  $x=0$  and spin reorientation temperatures for  $x=3, 6$ .
- (iii) The sign of curvature of the longitudinal magnetostriction curves changes from positive for the  $x=0$ , to negative in the  $x=3, 6$  due to conversion from the uniaxial to tilted magnetic anisotropy. At below transition temperatures, the value of the longitudinal magnetostriction at maximum applied field increases with the Co content and above  $T_{sr}$ , the saturation for  $x=6$  sample begins at lower field than for  $x=3$ .
- (iv) The transverse magnetostriction of  $\text{Tb}_3(\text{Fe}_{28-x}\text{Co}_x)\text{V}_{1.0}$  ( $x=0, 3, 6$ ) compounds exhibits a minimum at different applied fields that have been explained based on the anisotropy field in the  $b$ - $c$  plane of the unit cell. The minimum observed decreases with increase of Co content and temperature. In addition, the sign of curvature of transverse magnetostriction curves changes to negative for  $x=3, 6$  samples at above  $T_{sr}$ .

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