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A comparative study of the magnetoelastic properties of the $YFe_{10}V_2$ and $NdFe_{10}V_2$ compounds

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Abstract

The magnetoelastic properties of iron-rich $REFe_{10}V_2$ (RE = Nd, Y) compounds were studied via magnetostriction and thermal expansion measurements in the 5–300 K range of temperature in up to 6 T external fields. Results of thermal expansion analysis show that the spontaneous magnetostriction of the compounds mostly originates from itinerant magnetization. Besides, the small volume striction appearing in the thermal expansion of the Nd compound close to 50 K suggests the existence of a basal to conical spin re-orientation transition. The volume magnetostriction isotherms of both compounds take minimum values for external field corresponding to the anisotropy field. In addition, the anisotropic and the volume magnetostriction traces of the $NdFe_{10}V_2$ take marked maxima under low field, with a relatively large initial magnetostrictivity, again more pronounced at the conical–axial spin re-orientation transition ($T_{SR} = 130$ K). Analysis of the anisotropic magnetostriction of the Nd compound leads to the conclusion that the contribution of Nd–Fe interactions is negligible. The temperature dependence of volume magnetostriction is in good agreement with prediction of a phenomenological model based upon a fluctuating local band theory. This analysis shows that the difference between the forced volume strictions of Y and Nd compounds below and above T_{SR} originates from the Nd sublattice magnetization.

Keywords: Ferromagnetism; Thermal expansion; Magnetostriction; Magnetoelastic; Magneto-crystalline anisotropy

1. Introduction

Iron-rich $REFe_{12-x}M_x$ (1–12) alloys (where RE is a rare earth element, and M is V, Ti, Mo, etc…) have received great attention during past years as good candidates for hard permanent magnet applications. This comes from rather high Curie temperatures, excellent magnetic properties and a simple crystal structure [1,2]. The latter characteristic makes the series particularly appropriate for basic researches. The structure is tetragonal $ThMn_{12}$ type, SG I4/mmm, with three nonequivalent sites (8*i*, 8*j* and 8*f*) for Fe and M together and one site (2a) for rare earth. Compounds with nonmagnetic or light RE elements are ferromagnets and those with heavy RE elements are ferrimagnets. It was found that $Nd(Fe,V)_{12}$ compounds exhibit optimized intrinsic magnetic properties in the series [3–5]. In these (1–12)-type compounds, the Fe sublattice has uniaxial magnetic anisotropy, while that of the RE sublattice is planar. Competition of these two anisotropies potentially result in spin re-orientation transition T_{SR} from planar to axial and from low to high temperatures. For $NdFe_{10}V_2$, a spin re-orientation was found at $T_{SR}\sim130$ K [5].

Compared with other RE–Fe systems, only a few investigations on the magnetoelastic properties of the (1–12) series are reported to date [6–8]. Considering the nonmagnetic Y element, we have comparatively studied the magnetostriction and the thermal expansion of $REFe_{10}V_2$ with RE = Y, Nd, in order to question the contribution of local (4f) and itinerant (3d) magnetism on the magnetoelastic properties. This study is relevant for practical applications' as well as to better understand the basic magnetic behavior of Fe and Nd in 1–12 compounds.

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2. Experimental methods

Vanadium-containing compounds $Nd(Y)Fe_{10}V_2$ have been synthesized by arc melting of the constituent elements (grade purity 3 N) in a water-cold copper boat under high purified argon atmosphere. As-cast ingots were annealed in 10^{-4} mb evacuated chamber of a specific furnace. Annealing has been executed at $T = 960\,^{\circ}C$ for 24 h. The crystalline state of the compounds were assessed from X-ray diffraction (XRD) patterns on powdered samples. The patterns were recorded at room temperature (RT) using a computer controlled diffractometer with $Fe(K_\alpha)$ radiation and 0.05° step angle. The lattice parameter refinements have been done using the CELREF software. To check the orientation of the easy magnetization direction at RT, XRD analyses were performed on samples previously aligned in a field of 0.7 T parallel to the scattering vector.

For magnetization measurements, aligned cylindrical samples with 5 mm diameter were prepared by embedding the powders with epoxy resin and exposing until the epoxy hardened to a 0.7 T field parallel to the cylinder axis. Perpendicular alignments with respect to the cylinder axis were realized using a rotating device. Parallel magnetization measurements were performed at temperatures that comprised between 5 and 300 K and in magnetic fields up to 10 T, using an extraction-type magnetometer. The saturation magnetization was derived from the high-field extrapolation of M against $1/H^2$ curves to $1/H^2 = 0$. The anisotropy field H_a was deduced using the singular point detection (SPD) method. Temperature dependence of low-field magnetic susceptibility ($\chi(T)$) was measured using a high-sensitivity thermomagnetic torque. All our results of the magnetization of the samples were found as fairly consistent with the literature [3–5].

Magnetostriction and thermal expansion measurements were performed in the range 5–300 K and in up to 5 T external fields using semi-spherically shaped samples with ~6 mm diameter. For these measurements, a commercial capacitance dilatometer system was used. Magnetostriction was measured parallel (λ_t) and perpendicular (λ_n) to the field direction, from which the anisotropic magnetostriction, $\Delta\lambda = \lambda_t - \lambda_n$, and the volume magnetostriction, $\Delta v/v = \lambda_t + 2\lambda_n$, were deduced.

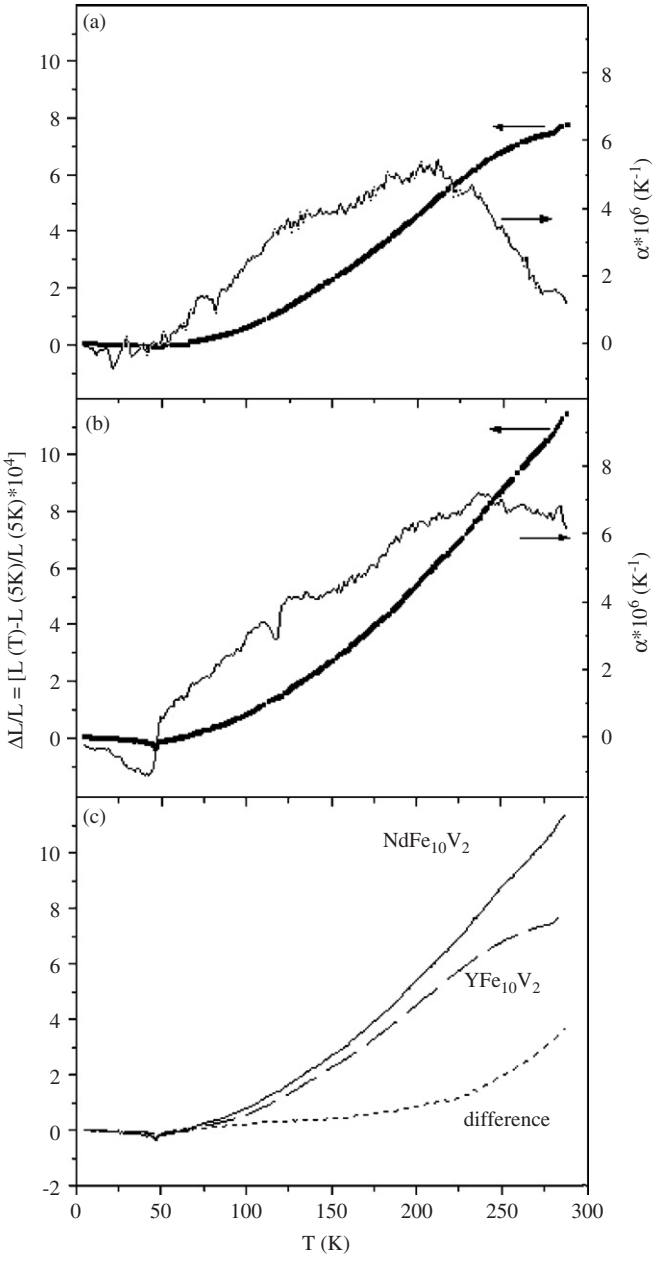

Fig. 1. Thermal expansion of (a) $YFe_{10}V_2$, and (b) $NdFe_{10}V_2$ and corresponding coefficients $\alpha \equiv (d/dT)(\Delta L/L)$. (c) Difference of the thermal expansion between the two studied compounds.

3. Results

X-ray diffraction patterns of the samples show that the samples are single 1–12 phase. Analysis of the patterns leads to the tetragonal lattice parameters $a = 8.56$ Å and $c = 4.77$ Å for $NdFe_{10}V_2$ alloy, and $a = 8.49$ Å and $c = 4.77$ Å for $YFe_{10}V_2$ alloy, in good agreement with the literature [1].

Fig. 1 shows the thermal expansion of both samples, as well as the differential expansion. The results are consistent with literature related to other 1–12 compounds, comprising magnetic and nonmagnetic RE, respectively [1,7]. In fact, the thermal expansion of both compounds should follow a T^4—Grüneisen-type behavior at low temperature and then it should exhibit an Invar-type behavior above RT. Although this is satisfied for the Y compound as shown in Fig. 1a, between 40 and 80 K the thermal expansion coefficient of the Nd compound does not fulfill the expected behavior [3], as shown in Fig. 1b. The most anomalous aspects of the results are first the small but negative thermal expansion coefficient of the $NdFe_{10}V_2$ compound as measured below 40 K and then a spin re-orientation type of transition occurring at ~50 K. Contrarily, the well-known spin re-orientation transition exhibited by $NdFe_{10}V_2$, from basal plan to c-axis and

setting at $T_{SR} = 130\,K$ [3,4], is accompanied by a weak expansion anomaly, only. However, this is consistent with previous measurements on the thermal expansion of $NdFe_{10.5}V_{1.5}$ we recorded using a strain gage method [9].

For both compounds, isothermal curves of the anisotropic magnetostriction as measured at selected temperatures are shown in Fig. 2. The magnetostriction traces of $NdFe_{10}V_2$ exhibit a different behavior than those recorded on $YFe_{10}V_2$. The anisotropic magnetostriction traces of $YFe_{10}V_2$ take a unique parabolic behavior for all temperatures. Contrarily, for $NdFe_{10}V_2$ an anomalous maximum is pointed out when applying weak fields and a sign reversal is evidenced when applying intermediate fields. As seen, both the amplitude and the characteristic field of the maximum depend on temperature. The peak is sharpened with a higher signal (for a larger initial magnetostrictivity) at the spin re-orientation temperature setting at $T_{SR} = 130\,K$. Comparison between Figs. 2a and 2b leads to consider two effects attached to Nd on the anisotropic magnetostriction. Firstly, Nd causes the onset of a spin re-orientation transition which corresponds to a local minimum in the magnetic anisotropy; as a consequence, this creates an anomalous maximum of magnetostriction in the low-field range. Secondly, the Nd contribution to the anisotropic magnetostriction is negative while that of Fe is positive. For $NdFe_{10}V_2$, competition of these two contributions is responsible for the observed magnetostriction compensation ($\Delta\lambda = 0$) taking place in the intermediate range of fields as shown in Fig. 2b.

Fig. 3 shows the volume magnetostriction of $NdFe_{10}V_2$ and $YFe_{10}V_2$ plotted versus the applied fields for selected temperatures. In the case of $YFe_{10}V_2$, the volume magnetostriction is nearly zero under weak fields and then it passes through a minimum for higher fields ($H = H_{cr}^{Y}$), also depending on temperature as shown in Fig. 3a. Since the applied field ranges close to the magnetic anisotropy field of $YFe_{10}V_2$ [10], the observed minimum indicates that the collective rotation of Fe moments toward the applied

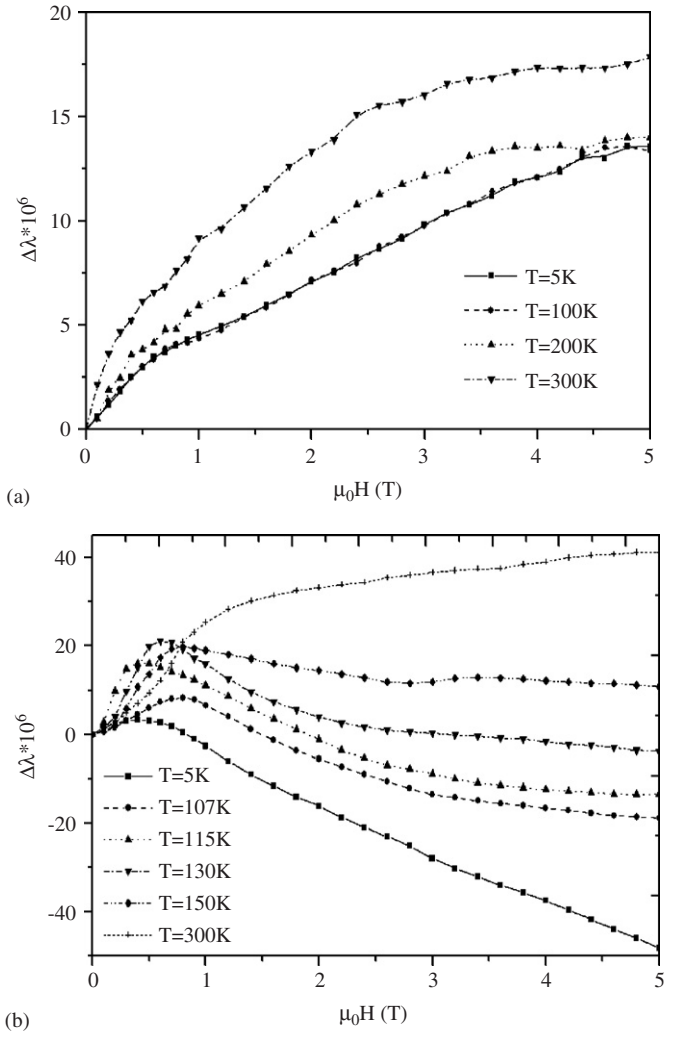

Fig. 2. Isothermal curves of the anisotropic magnetostriction of (a) $YFe_{10}V_2$ and (b) $NdFe_{10}V_2$ compounds as a function of applied field at typical temperatures.

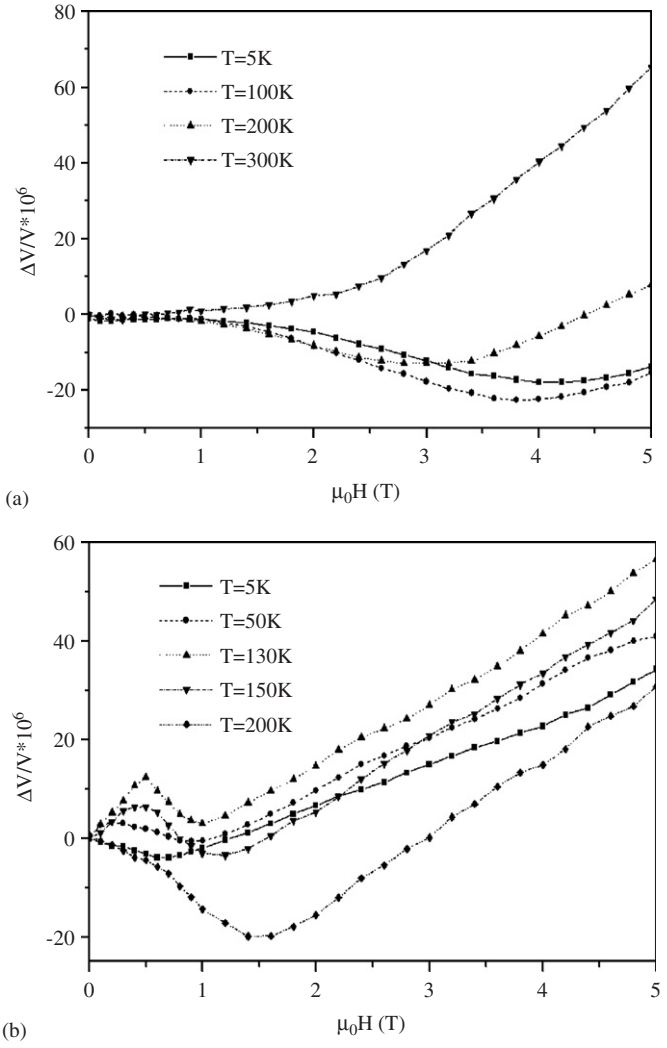

Fig. 3. Isothermal curves of the volume magnetostriction of (a) $YFe_{10}V_2$ and (b) $NdFe_{10}V_2$ compounds as a function of applied field at typical temperatures.

field direction is accompanied by a positive volume striction. Also for different applied fields ($H = H_{cr}^{Nd} \neq H_{cr}^{Y}$), the traces of volume magnetostriction of $NdFe_{10}V_2$ exhibit similar minima as shown in Fig. 3b. Moreover, the traces reveal a maximum under lower fields ($H = H_{max} < H_{cr}^{Nd}$), which is more pronounced when temperature tends toward T_{SR}. Such behavior was also observed for $TbFe_{11}Ti$ in the vicinity of a corresponding spin re-orientation transition [11]. The maxima are similar to those appearing with the anisotropic magnetostriction, having probably a unique origin, e.g. a minimum of the local magnetic anisotropy at spin re-orientation transition.

4. Discussion

The difference between the two thermal expansion traces of Fig. 1c allows to point out a thermoelastic effect related to the Nd–Nd and Nd–Fe interactions. Then, comparison of the thermal expansion measured parallel on both the Y and Nd compounds demonstrated that the Nd sublattice induces two clear impacts on the thermal expansion curves. Firstly, there is a small negative thermal expansion coefficient for $NdFe_{10}V_2$ below 40 K and a spin re-orientation-type transition at about 50 K, which do not exist on the thermal expansion traces of $YFe_{10}V_2$. Secondly, the characteristic negative curvature taking place close to RT in the thermal expansion of $YFe_{10}V_2$ is weakened in the case of $NdFe_{10}V_2$. To the latter phenomenon corresponds a positive magnetovolume effect. This isotropic contribution may be related to the larger $\langle$Fe–Fe$\rangle$ distances found for $NdFe_{10}V_2$ than for $YFe_{10}V_2$, especially between the Fe(8*f*) atoms, which leads to the experimental increase by ~10% of the Fe magnetic moments, according to the Bethe–Slater curve [3,4,12]. Also, the exchange interactions acting between the Fe atoms, which are responsible for the isotropic spontaneous volume magnetostriction, should be enforced. A higher Curie temperature of $NdFe_{10}V_2$ than of $YNd_{10}V_2$ reveals consistency with increases of both the Fe moments and the Fe–Fe interactions. Otherwise, considering the weak-anomalous behavior of the thermal expansion in the spin re-orientation region, the contribution of the Nd–Fe exchange interactions to the thermal expansion should be neglected.

The unexpected exhibition in $NdFe_{10}V_2$ of a spin re-orientation type transition at T~40 K could be attributed to the effect of higher order crystal field parameters [13]. Comparison made with $DyFe_{11}Ti$, in which Dy like Nd has negative first and second order Stevens parameters (α, $\beta < 0$), may assume that the specific plan-to-axis transition of the easy magnetization direction of $NdFe_{10}V_2$ could take place in a two-step phenomenon [2,14]. The low-temperature magnetic arrangement should change via a sharp first order transition from basal plan to cone magnetic structure at T~40 K, then it could transform gradually (second order transition) from cone to the *c*-axis collinear structure existing at higher temperatures [1]. This analysis must be confirmed using both neutron diffraction and magnetization measurements.

To compare the anisotropic magnetostriction of the Y and Nd-based compounds, the corresponding isothermal traces of the $YFe_{10}V_2$ sample were fitted and scaled, accounting for the reduced magnetization ($m(T,H) = M(T,H)/M(T,H \to \infty)$). Typically for 5 and 300 K and as represented in Fig. 4, our analysis shows good consistency between the isotherms of the anisotropic magnetostriction ($\Delta\lambda_{exp}$) and those of the reduced magnetization in the form of the following scaling relation:

$$\Delta\lambda_{exp}(m) = a_0 + a_1 m^2(T,H) + a_2 m^4(T,H) + a_3 m^6(T,H). \quad (1)$$

The adjustable coefficients a_i were calculated at different temperatures by using a least-squares fit method, whose values are reported in Fig. 4, for two typical temperatures. Accounting for the nonmagnetic nature of Y, one may relate this specific behavior to Fe sublattice contribution to the anisotropic magnetostriction, also valid for all RE-$Fe_{10}V_2$ compounds. Thus, for $NdFe_{10}V_2$, where the magnetic anisotropy of the Fe sublattice is dominant above T_{SR}, it is worth assuming the same functional dependence for the isothermal curves of anisotropic magnetostriction around RT.

However, at this point, a peculiar consideration on the anisotropic magnetostriction term of $NdFe_{10}V_2$ sample can be made. From Fig. 2, it is clear that both series of curves (Y and Nd compounds) exhibit similar behaviors, except for the initial slopes (magnetostrictivities) and for the saturation values, differences that can be attributed to the magnetic anisotropy of Nd atoms and the different

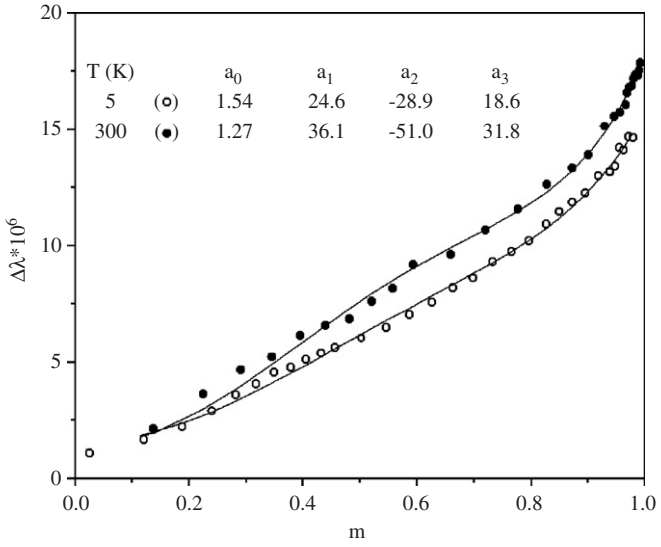

Fig. 4. Comparison of the experimental data (symbols) for $YFe_{10}V_2$ compound with the scaling form of Eq. (1) (continues lines), typically at 5 and 300 K temperatures. The least square fitted values of the adjustable coefficients, a_is, at 5 and 300 K are given.

saturation magnetization of the two compounds, respectively. So, we propose to express the isothermal curves of anisotropic magnetostriction of $NdFe_{10}V_2$ as a combination of the 5 and 300 K isothermal curves (i.e. $\Delta\lambda(5,H)$ and $\Delta\lambda(300,H)$, respectively), in the form of

$$\Delta\lambda(T,H) = D(T)\Delta\lambda(5,H) + E(T)\Delta\lambda(300,H), \quad (2)$$

where $D(T)$ and $E(T)$ are adjustable coefficients referring to the 5 and 300 K contributions, respectively. In this way, it is possible to distinguish a contribution of itinerant magnetism type (Fe sublattice) as the second term, from that of localized magnetism type (Nd moment) as the first term. Based on such derivation and using a least-squares fit method, values of the $D(T)$ and $E(T)$ coefficients were calculated versus temperature as shown in Fig. 5. It is clear that the Fe contribution ($E(T)$) rapidly increases around T_{SR}, and the Nd contribution ($D(T)$) gradually decreases upon increasing temperature, then exhibiting a clear but limited jump at $T\sim115$ K, very close to T_{SR}.

In agreement with the above mentioned assumption, we find a good consistency between the Fe contribution to the anisotropic magnetostriction of $NdFe_{10}V_2$ ($E(T)\Delta\lambda(300,H)$) and the scaling form of Eq. (1) with the appropriate a_i coefficients. This confirms the negligible magnetoelastic contributions of the Nd–Fe interactions.

Also, a comparison of Figs. 3a and b shows that $H_{cr}^{Nd} < H_{cr}^{Y}$. This can be due to the lower relative magnetic anisotropy of $NdFe_{10}V_2$ in comparison with that of $YFe_{10}V_2$ [15]. Additionally, Nd has marked impact on the magnitude of volume magnetostriction, especially below and close to T_{SR}. The specific impact of Nd can be derived from the temperature dependence of the volume magnetostriction under low ($\mu_0 H = 0.1$ T) and high ($\mu_0 H = 5$ T) fields as reported in Fig. 6, and can be explained by a phenomenological theory based on a fluctuating band model, proposed by Shiga [16]. In the theoretical derivation, both itinerant electron and local moments were accounted for, so that the volume magnetostriction was described as

$$\Delta V/V = a\chi(T)M(T) + bM^2(T) + \text{constant}, \quad (3)$$

where a and b are coefficients containing the magnetovolume coupling constant and the compressibility, and $\chi(T)$ and $M(T)$ are the temperature-dependent susceptibility and the bulk magnetization, respectively. The first term of Eq. (3) is a band term and refers to the contribution of itinerant electrons, while the second term concerns the contribution of interactions as described by a localized model. Thus, we deduce $\Delta V/V = 142.4\,\chi(T)M(T) - 0.39M^2(T) + 100.5$ as a master equation, in good agreement with the experimental data obtained at 5 T, as shown in Fig. 6b. For comparison, simulated data are plotted with open circles in Fig. 6b. Once more, it is worth underlining that the local magnetization of the Nd sublattice is made first responsible for the

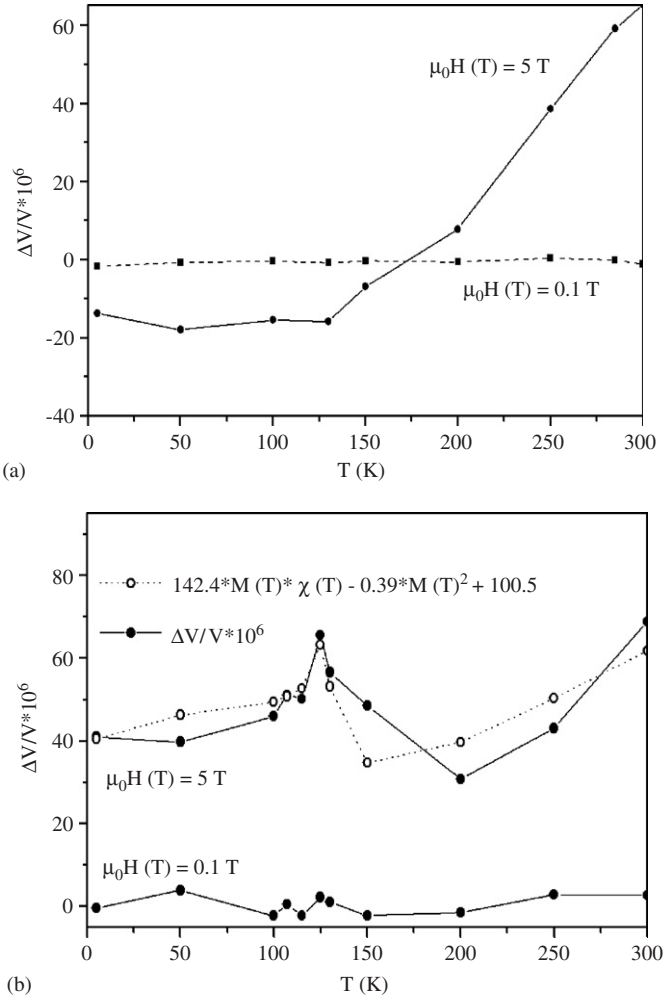

Fig. 6. Temperature dependence of the volume magnetostriction of (a) $YFe_{10}V_2$ and (b) $NdFe_{10}V_2$ compounds at weak ($\mu_0 H = 0.1$ T) and high ($\mu_0 H = 5$ T) fields.

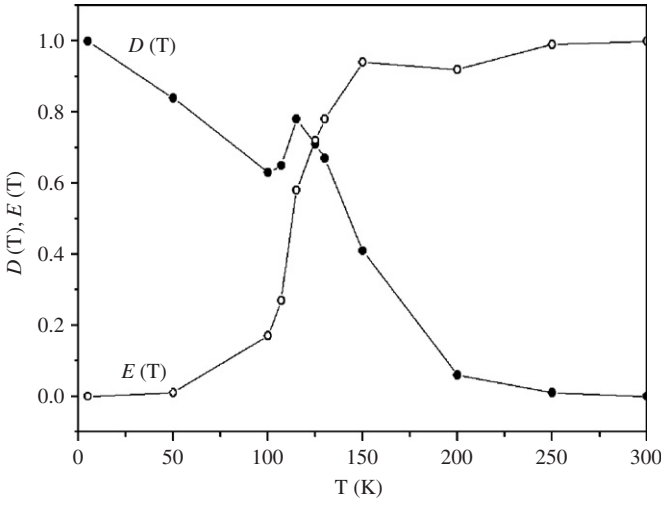

Fig. 5. Temperature dependence of $D(T)$ and $E(T)$ coefficients, deduced after fitting of the isothermal curves of anisotropic magnetostriction of $NdFe_{10}V_2$ compound with Eq. (2). $D(T)$ and $E(T)$ refer to the local and itinerant contributions to the anisotropic magnetostriction of $NdFe_{10}V_2$ compound, respectively.

distinguished temperature dependences of the Y and Nd volume magnetostriction behavior below and around T_{SR}.

5. Conclusions

The magnetoelastic properties of $YFe_{10}V_2$ and $NdFe_{10}V_2$ compounds were comparatively studied via magnetostriction and thermal expansion measurements. Results show that:

(1) The Nd sublattice leads to two main effects on the thermal expansion, reference to the Y compound. The first consists in a small negative thermal expansion coefficient below 40 K and a spin re-orientation-type transition at ~50 K, and the second consists in a positive volume striction at RT. The first effect can be correlated to a first-order basal-conical spin re-orientation transition, and the second one originates from enforced exchange interactions taking place in $NdFe_{10}V_2$.
(2) The anisotropic magnetostriction of $NdFe_{10}V_2$ contains two contributions. The first contribution, which is more pronounced below T_{SR}, results directly from the Nd magnetization. The second part, which is mainly responsible for the magnetostriction observed close to RT, is related to the itinerant-type magnetization of Fe. Thirdly, it appears that the Nd–Fe interactions do not contribute to magnetostriction. Competition of the two former contributions creates a magnetostriction compensation under intermediate fields.
(3) Isothermal traces of the $NdFe_{10}V_2$ anisotropic magnetostriction exhibit transient maxima under low fields that appear more accentuated for temperature approaching T_{SR}. This could result from a related minimum of magnetic anisotropy close to T_{SR}. The same behaviors having the same origin appears in the isothermal traces of volume magnetostriction.
(4) Isothermal traces of volume magnetostriction of both compounds exhibit wide minima for intermediate fields, more specifically when the applied field is equivalent to the magnetic anisotropy field. In fact, closeness of these fields for $YFe_{10}V_2$ leads that the coherent rotation of the Fe moments towards the applied field is accompanied by a positive volume striction.
(5) Reference to a local band model, shows that the different temperature dependence of the volume magnetostriction at saturation for the Nd and Y compounds below and up to T_{SR} originates directly from the Nd sublattice magnetization.

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