

Magnetostriction and First Order Magnetic Transition in $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ Compounds

N. Tajabor*, M.R. Alinejad¹ and F. Pourarian²

In this paper, the effect of partial replacement of Nd by Dy atoms on the magneto-elastic properties of $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ (2-14-1 type) alloys with $x = 0.1, 0.3$ and 0.5 , in a temperature range of 77 K to room temperature have been studied using magnetostriction measurement. The first Order Magnetic Process, FOMP, was determined by measuring the temperature dependence of low field magnetic susceptibility. Results show that an indicative temperature for FOMP, T_{SP} , increases about 40 K with increasing Dy content, which can be due to the non-negligible role of the higher than second order terms ($\kappa_4^m, \kappa_6^m, \dots$) of the crystalline field anisotropy of the rare earth sub-lattice. This conclusion is confirmed via magnetostriction results, which show that the partial substitution of Dy for Nd increases the anisotropic magnetostriction. This is attributed to a stronger magneto-crystalline anisotropy. At $T > T_{SP}$, Dy partial substitution also causes an increase in the volume magnetostriction, which indicates a positive exchange magnetostrictive effect of Dy - Fe anti-ferromagnetic interaction.

INTRODUCTION

There have been at least two reasons to emphasize the study of the magnetic properties of $\text{Nd}_2\text{Fe}_{14}\text{C}$ compound, which are isostructure with the familiar $\text{Nd}_2\text{Fe}_{14}\text{B}$ compounds. First, $\text{Nd}_2\text{Fe}_{14}\text{C}$ compounds have the advantage of solid-state transformations, which can be used to obtain magnets with high bulk coercivity without using powder metallurgy or rapid solidification methods, i.e. directly from the cast structures [1,2]. Second, the magnetic anisotropy field in the carbides is larger than the boride compounds. This is partly attributed to the electric field of the negative interstitial C ions acting on Nd atoms [3,4].

Preparation of $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ solid solutions is possible for a full range of x values ($0 \leq x \leq 2$) [5-7]. All of these compounds crystallize in the tetragonal structure with space group $\text{P4}_2/\text{mnm}$, in which rare earth atoms occupy two nonequivalent sites 4f and 4g [8]. According to [9], partial substitution of Nd by Dy enhances the anisotropy field and, also, the

coercivity, as expected, due to the higher anisotropy field of Dy compared to Nd. However, the saturation magnetization of the substituted samples decreases with the increase of Dy content. This is due to the effect of anti-parallel alignment of Dy and Fe magnetic moments [10,11]. Therefore, the $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds with small values of the substituted x are mostly interesting for use in technological applications.

Both spin re-orientation and First Order Magnetic Process (FOMP), have been observed in this compound [10,12]. Spin re-orientation means that the easy magnetization direction changes from one crystallographic direction to another with temperature change. In $\text{Nd}_2\text{Fe}_{14}\text{C}$ compounds, this kind of transition is in the form of easy axis rotation from canted to tetragonal c -axis with increasing temperatures beyond $T_{SR} \cong 135$ K. The spin re-orientation temperature, T_{SR} , decreases when Dy is partially substituted for Nd [13,14]. The first order magnetization process is defined as the discontinuities or jumps exist in the magnetization curve, when sufficient high external magnetic fields are applied along a special crystallographic direction. In the $\text{Nd}_2\text{Fe}_{14}\text{B}$ compound, it has been found that a FOMP can only be observed below $T \cong 200$ K and in the presence of external fields higher than $H \cong 15^\circ\text{T}$ applied parallel to a [100] crystallographic direction [15]. In the case of $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$, experimentally obtained field dependent magnetization curves show that the field dependence of FOMP is almost independent

*. Corresponding Author, Department of Physics, Faculty of Sciences, Ferdowsi University of Mashad, Mashad, I.R. Iran.

1. Department of Physics, Faculty of Sciences, Ferdowsi University of Mashad, Mashad, I.R. Iran.
2. Carnegie Mellon Institute, Carnegie Mellon University, Pittsburgh, Pa 15219, USA.

of the Dy content for $x \leq 0.4$ [10]. However, other researcher's work has indicated that all coefficients of the magneto crystalline anisotropy energy change upon partial substitution of Dy for Nd [4,16-18]. For the $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds, an attempt was made to examine the effect of partial substitution of Dy for Nd on the temperature dependence of FOMP and on the magnetostrictive properties. According to the ac-magnetic susceptibility results on $\text{Nd}_2\text{Fe}_{14}\text{C}$ [12], one expects a kind of cusp like behavior correlated to the FOMP transition in low field ac-magnetic susceptibility of this compound. Based on this, therefore, the ac-susceptibility and magnetostriction of $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ with $x = 0.1, 0.3$ and 0.5 compounds were investigated. Below, a possible unified description will be provided for both spin re-orientation and FOMP transitions, based on the magnetic anisotropy energy considerations.

THEORETICAL ASPECTS

It is a fact that for the rare earth-iron (RE-Fe) compounds, the magnetic anisotropy is mostly introduced by crystal field effects acting on $4f$ -electrons of RE atoms. The crystal electric field energy, H_{CEF} , in terms of Steven's operators, O_n^m , is written as [19]:

$$H_{\text{CEF}} = \sum_{n=2,4,\dots}^{\infty} \sum_{m=0}^n \theta_n \langle r_{4f}^n \rangle A_n^m O_n^m, \quad (1)$$

The Steven's operators, O_n^m , as symmetric polynomials constructed from components of the total momentum, J , of RE atoms, are appropriately provided as a base for symmetric expansion of H_{CEF} , in each crystal structure. With the modified notation of $\theta_2 \equiv \alpha_J, \theta_4 \equiv \beta_J, \theta_6 \equiv \gamma_J, n$ is the appropriate Steven's factor of n order that represents the type of asphericity of the $4f$ -charge distribution. The coefficients A_n^m , which refer to crystal field coefficients, describe the electrostatic potential of the charge surrounding the $4f$ -electrons. Values for $\langle r_{4f}^n \rangle$, where $\langle r_{4f}^n \rangle$ is the mean value of the n order power of the $4f$ -radius, are the multi-pole charge moments of the $4f$ -shell.

Alternatively, the anisotropy energy is almost approximated by some terms of the following phenomenological expansion:

$$E_a = \sum_{n=2,4,\dots}^{\infty} \sum_{m=0}^n \kappa_n^m P_n^m(\cos \theta) \times \cos m\phi. \quad (2)$$

The angles θ and ϕ are the polar and azimuthal angles of magnetization with respect to the main symmetry directions of the unit cell (a, b and c for tetragonal, for example). P_n^m is a familiar Legendre function; and κ_n^m is the phenomenological temperature dependent anisotropy coefficient. Each term in this expansion

will be minimum for special values of θ and ϕ . The easy direction of magnetization at each temperature will be determined by competition between various terms to make E_a minimum. In the case of RE-Fe compounds, where magnetic anisotropy is mainly the magneto-crystalline type, Equation 2 can be considered as a phenomenological description of the microscopic expansion of CEF energy (Equation 1). So, one can define the following relation between anisotropy coefficients and thermodynamic average of Steven's operators:

$$\kappa_n^0 \equiv \theta_n \langle r_{4f}^n \rangle A_n^0 \langle O_n^0 \rangle, \quad n = 2, 4, 6, \quad (3)$$

and some similar relations for necessary higher order terms. Using this terminology one can say that the temperature and field dependence of anisotropy energy (particularly κ_n^m) are the intrinsic origins of both spin re-orientation and FOMP transitions. These two transitions are associated with the fact that there is more than one non-equivalent minimum in the magnetic free energy surface. For example, if one plots total anisotropy and Zeeman energy, due to the external magnetic field of the $\text{Nd}_2\text{Fe}_{14}\text{C}$ compound, with respect to the angle between magnetization and c -axis at temperatures lower than $T \cong 200$ K, it will show two minima according to [100] and [001] crystallographic directions, in addition to another one around a 30° angle, according to the equilibrium state of the easy magnetization direction at $T < T_{SR}$ temperatures. The relative thermal and field stability of these minima can be described based on the temperature and field dependence of the second, fourth and sixth order terms ($\kappa_2^0, \kappa_4^0, \kappa_6^0$) in Equation 2. Therefore, the nature of the FOMP is similar to that of the spin re-orientation. In other words, a spin re-orientation can be considered as a zero field FOMP.

Within the above single ion approximation, the temperature dependence of magnetostriction, λ , can be described in terms of the reduced hyperbolic Bessel functions, I , after Cullen's classical theory [20,21]:

$$\lambda = \sum_{l=2,4,\dots} \lambda_l \times I_{2l+1/2}(Y), \quad (4)$$

where λ_l 's are experimentally adjustable magneto-elasticity coefficients and Y is obtained from the following relation in terms of reduced magnetization, m , of the RE sub-lattice:

$$m(T) \equiv \frac{m_s(T)}{m_s(0)} = I_{3/2}(Y) = \text{cotgh}(Y) - \frac{1}{Y}. \quad (5)$$

Here, $m_s(0)$ and $m_s(T)$ are magnetization of rare-earth (RE) sub-lattice at zero and T temperature, respectively. Using the above unified terminology for single ion effects on magnetic anisotropy, one can

approximately consider each term in magnetostriction expansion, Equation 4, parallel with similar terms in Equations 1 and 2. Actually, within a single-ion model, in order to calculate the magnetoelastic coupling, the constants in Equation 4 ($\lambda_2, \lambda_4, \lambda_6$), will be proportional to Steven's reduced matrix elements n (multipoles), if one assumes that α_J, β_J and γ_J are parallel with $l = 2, l = 4$ and $l = 6$, respectively. Although the second order term in Equation 4, ($I_{5/2}$), effectively describes the consistency of the magnetostriction results with Callen's theory, variations of relatively small fourth and sixth order terms with composition are interesting enough to be considered, in order to obtain some idea of similar variations in parallel terms in anisotropy and CEF energies. For this purpose, Cullen's model, despite its simplicity, is too flexible to simulate experimental data with a straightforward fitting routine [22]. Sometimes, the following scaling expansion, based on the low temperature behavior of hyperbolic Bessel functions, has been used [18,20-22]:

$$\Delta\lambda(T) \equiv \lambda_{02}m^3(T) + \lambda_{04}m^{10}(T) + \lambda_{06}m^{21}(T). \quad (6)$$

In which λ_{0n} 's, once again, are modified magnetoelasticity coefficients.

Bellow, this terminology will be used more to describe the experimental results on $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds.

EXPERIMENTAL PROCEDURE

$\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds, with $x = 0.1, 0.3$ and 0.5 , were prepared by RF induction melting of the stoichiometric mixed powder of the constituent elements. Ingots were annealed under Argon atmosphere for more than one week at 880°C [23]. XRD patterns showed that all three samples were nearly single phase, the amount of impurities such as $\alpha\text{-Fe}$ and 2-17 phases being less than about 5%. Scanning electron microscopy studies also revealed a uniform distribution of polycrystalline grains of a few microns in the samples.

Magnetostriction measurements were performed on disc shaped samples with 5 mm diameter and 1.5 mm thickness. For these measurements, a strain gauge of type SK-09-031-DE350 was mounted on the flat surface of each sample. The relative change in electric resistance of these gauges is equal to 2 times the produced strain in them ($\Delta R/R = 2 \cdot \Delta l/l$). Application of a magnetic field to a magnetostrictive sample causes the dimension of the sample to change. By this change, the mounted gauge on the sample undergoes strain and, therefore, its electric resistance changes. Relative change of resistance is measured by a Wheatstone bridge with a sensitivity in the order of

10^{-7} and then the amount of strain in the sample is obtained. The measuring circuit is shown in Figure 1. The dummy gauge, R_d , which is mounted on a copper disc, is to eliminate the unwanted bridge output, due to change of gauge resistance with temperature or field [24].

After applying a magnetic field parallel to the flat surface of the sample, the magnetostriction parallel ($\lambda_{||}$) and normal (λ_{\perp}) to the field direction was measured, then, anisotropic magnetostriction, $\Delta\lambda = \lambda_{||} - \lambda_{\perp}$, and volume magnetostriction, $\Delta v/v = \lambda_{||} + 2\lambda_{\perp}$, were calculated [25]. Based on circular symmetry of the flat surface of the samples, the demagnetization or form effects are equal in measured $\lambda_{||}$ and λ_{\perp} , so are canceled in anisotropic magnetostriction. The quantity, $\Delta\lambda$, is a representative of magnetostriction in a constant volume and it is free from an initial demagnetized state. In the case of isotropic polycrystalline samples with crystallites of tetragonal symmetry, they can be described, up to the second order magnetoelastic terms, by [26]:

$$\Delta\lambda \propto (\lambda_2^{\alpha,2} + \lambda^{\gamma,2} + 2\lambda^{\epsilon,2}), \quad (7)$$

where $\lambda_i^{j,k}(H, T)$, the phenomenological magnetostriction constants, have their usual meanings, ($\lambda_2^{\alpha,2}$) is the $\frac{c}{a}$ ratio distortion, ($\lambda^{\gamma,2}$) is the shear breaking of the basal plane cylindrical symmetry, and ($\lambda^{\epsilon,2}$) is the shear tilting of the c -axis. In the same isotropic polycrystalline sample, the volume magnetostriction can be described by:

$$\omega \propto \lambda_1^{\alpha,0}, \quad (8)$$

where $\lambda_1^{\alpha,0}$ constant presents the isotropic part of magnetostriction and does not depend on the magnetization direction, therefore, it does not reduce the lattice symmetry. This means that the volume magnetostriction is only contributed to by

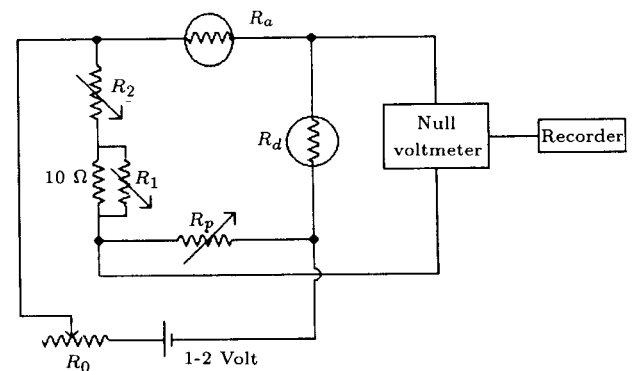


Figure 1. Schematic of the circuit for measuring magnetostriction. R_a is an active strain gauge mounted on the sample and R_d is the dummy gauge mounted on a copper disc.

the uniform exchange-striction mode, which can be separated out of anisotropic parts in Equation 7, even with measurements on a polycrystalline sample [18].

Magnetic ac-susceptibility of the samples, which is the response of magnetization to the change in the external magnetic field, was also obtained using an ac-susceptometer 7000, Lake-Shore. In this system, an ac-magnetic fields with frequencies between zero up to 1000 Hz can be employed.

RESULTS AND DISCUSSIONS

It is expected that at a particular temperature, where the two spin phases and, so, their respective minima in free energy, have nearly equal probability, the anisotropy field goes through a minimum and magnetic susceptibility tends to maximum. Two peaks or cusps have been observed in the real part of the magnetic ac-susceptibility versus temperature of $\text{Nd}_2\text{Fe}_{14}\text{C}$ compounds by Kou et al. [12]. The peak at lower temperature ($T = T_{SR}$) is explained by the spin re-orientation from the easy cone to the c -axis. The second cusp at higher temperature ($T = T_{SP}$) is attributed to type-1 FOMP from [001] to [100] direction. Figure 2a shows a similarity to the later cusp in the presented results on temperature dependence of ac-susceptibility for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ solid solutions. Temperatures corresponding to the maximum of the curves, (T_{SP}), are plotted versus x in Figure 2b. Expected first peaks at T_{SR} should be translated to lower than 120 K temperature, according to the usual decreasing behavior of T_{SR} with Dy content, as shown in Figure 3a [10]. From Figure 2b, it is evident that the partial substitution of Dy causes an increase in T_{SP} . Therefore, one can introduce a FOMP transition

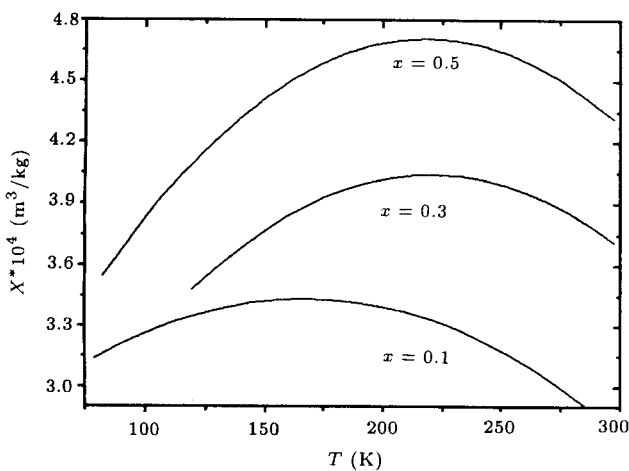


Figure 2a. Ac-magnetic susceptibility as a function of temperature for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ samples, with $x = 0.1, 0.3$ and 0.5 , at similar conditions (Ac-field = 50 A/m, dc-field = 0, $f = 333$ Hz, Temp. rate = -3 k/min).

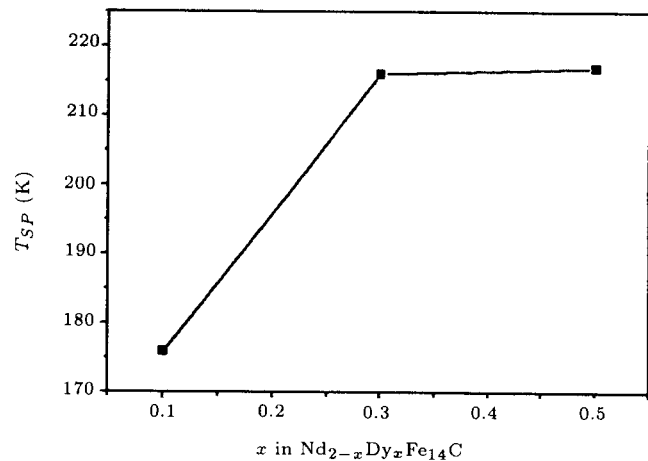


Figure 2b. The temperature according to the maximums in ac-magnetic susceptibility curves (T_{SP}) as a function of Dy content in $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$.

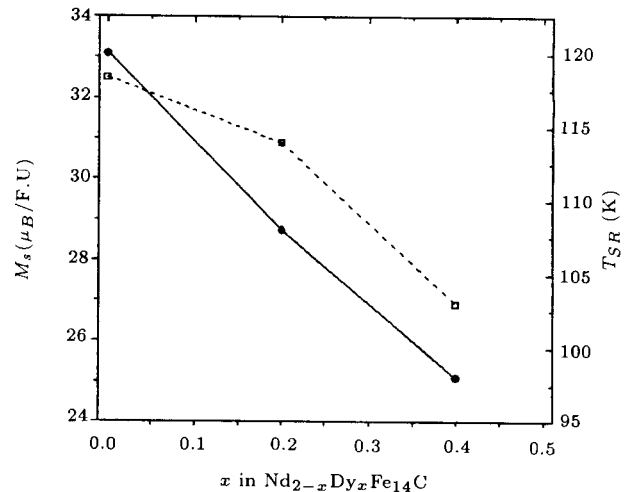


Figure 3a. Saturation magnetization (M_s) and spin re-orientation temperature (T_{SR}) as a function of Dy content in $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{B}$ compounds [10].

at higher temperatures by substitution of Nd by Dy atoms. This means that the possibility of having a minimum in anisotropy in the [100] direction increases by partial replacement of Nd by Dy. It is clear that the stability of the [100] minimum can be larger than the [001] only in the presence of large external magnetic fields ($H > 15\text{T}$) applied in the [100] direction. This indicates that, although the second order anisotropy term in anisotropy energy (first acceptable term in Equation 2 based on symmetry considerations, κ_2^0), which is responsible for [001] axial anisotropy at these temperatures, has a dominant role, the higher order terms, $\kappa_4^m, \kappa_6^m, \dots$, are not negligible. Previously, a similar beneficial effect for the role of the Dy content on the fourth anisotropy parameter has been considered by Kim et al. [14] using magnetization measurements. Figure 3b shows their results for K_1, K_2 anisotropy constants, which are proportional to κ_2^0, κ_4^0 , anisotropy

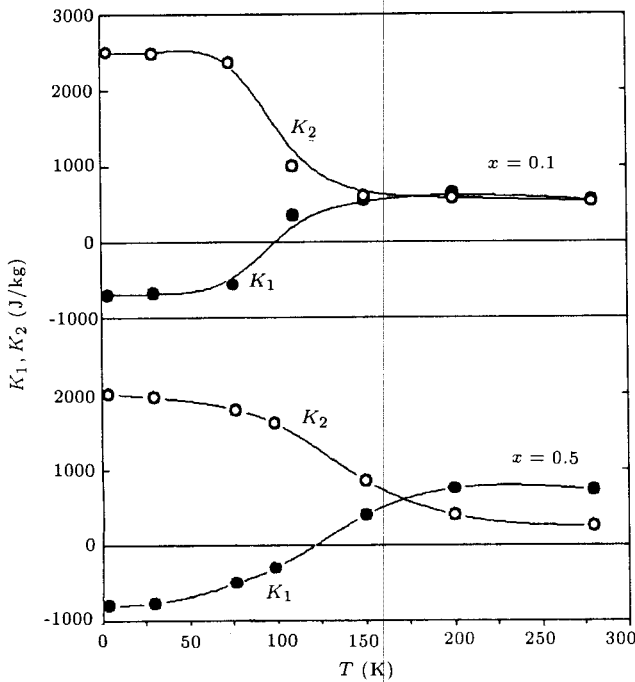


Figure 3b. Temperature dependence of first and second order anisotropy constants for $x = 0.5$ and $x = 1$ in $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{B}$ [14].

coefficients, respectively. Below, this effect will be studied in more detail. In addition, the observed saturation behavior of T_{SR} for $x > 0.3$ in Figure 3b can be attributed to the higher decrease in κ_4^m, κ_6^m anisotropy coefficients than κ_2^0 with increasing temperature.

Linear magnetostriction values, in a constant external field of 1.4 T and in the selected temperatures ranging from 77 K to room temperature, are shown in Figure 4a. It can be seen that with increasing Dy content, overall values of the linear magnetostriction are increased, which can be due to an increase in the magneto-crystalline anisotropy. These curves also show that in the temperature range 150 to 220 K, magnetostriction values decrease, which is due to the mentioned temporarily minimizing of magnetic anisotropy around T_{SP} . At temperatures above FOMP region and near to the Curie temperature ($\cong 535$ K), magnetization of the sample starts to decrease due to thermal agitation. Therefore, it is expected that with an increase in temperature and in passing through the above mentioned temperature range, the increasing magnetostriction trend in Figure 4a, ends and magnetostriction curves start to decline.

The anisotropy minimization has a considerable effect on the normal component of the magnetostriction. Figure 4b shows that normal magnetostriction becomes zero in the T_{SP} region, while its magnitude increases with Dy content. As a consequence, the anisotropic magnetostriction changes with temperature

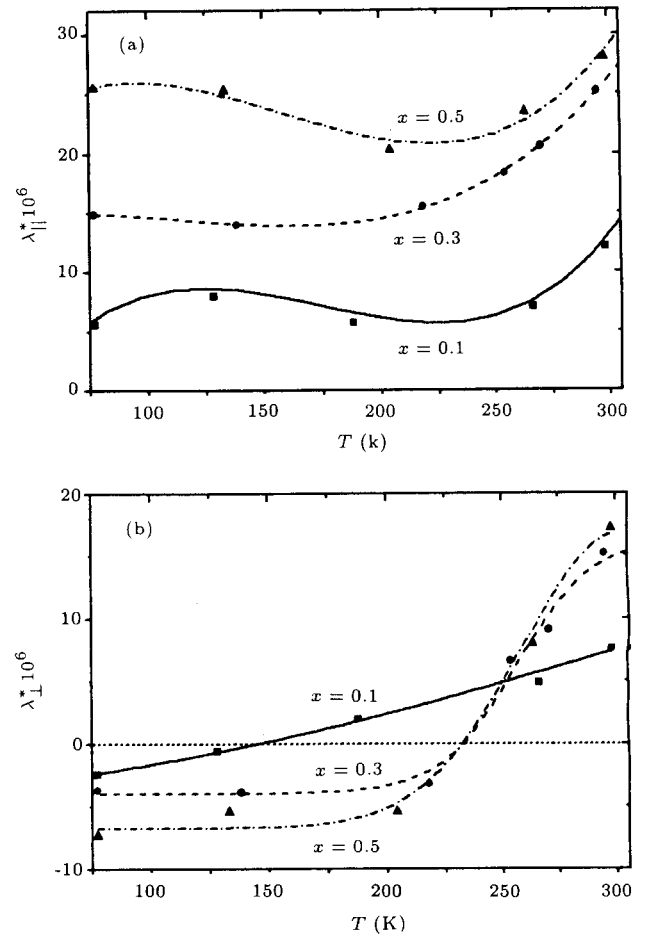


Figure 4. Linear magnetostriction parallel (a) and perpendicular (b) to the magnetic field direction at constant field of $H = 1.4$ T for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ samples, with $x = 0.1, 0.3$ and 0.5 . The curves are for eye guide.

as shown in Figure 5. Within the limits of experimental accuracy ($\cong 4 \times 10^{-6}$), it is possible to show that the temperature dependence of magnetostriction is consistent with the second order Callen's single particle theory (first term in Equation 4):

$$\Delta\lambda = \lambda_2 \times I_{5/2}(Y). \quad (9)$$

By assuming that the temperature dependence of the magnetization of the RE sub-lattice does not change appreciably by the partial replacement of Nd by Dy, then it is possible to use magnetization results of the RE sub-lattice in the original composition ($x = 0$) [27], for the samples presented here and simulate the temperature dependence of magnetostriction in Figure 5 from Equations 5 and 9. In order to apply this approximation, two of the following facts for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds were considered. First, partial substitution of Dy atoms does not affect the temperature dependence of magnetization considerably, as it seems in Figure 6. Actually, this substitution yields a permanent magnet with a temperature behavior comparable with the original composition [2].

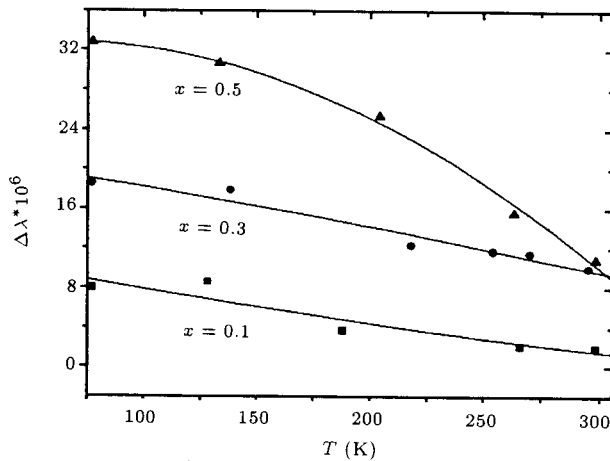


Figure 5. Anisotropic magnetostriction at selected temperatures shown with symbols for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ samples, with $x = 0.1, 0.3$ and 0.5 in $H = 1.4 \text{ T}$.

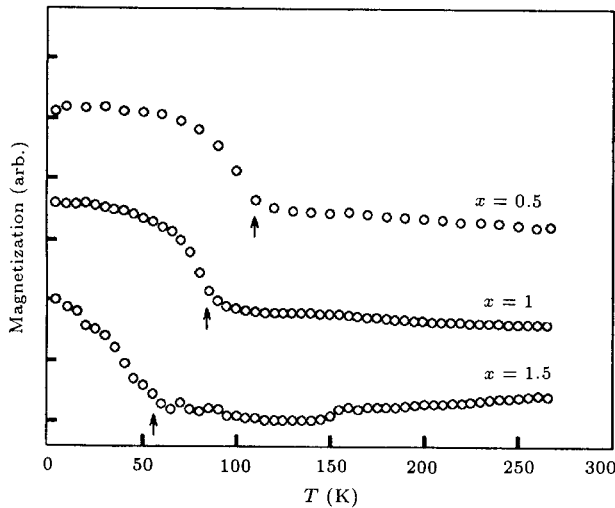


Figure 6. Temperature dependence of magnetization of $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{B}$ measured perpendicular to the alignment direction in applied fields below 3.2 kA/m . Arrows indicates spin re-orientation temperatures [14].

Secondly, only about 30% of the magnetization of these compounds should be introduced by the RE sub-lattice and the rest of it originates from iron sub-lattice [2,27,28]. In each case, more than 15% error in the results is not expected with this approximation. Curves obtained by choosing $\lambda_2(x = 0.1) = 10$, $\lambda_2(x = 0.3) = 25$ and $\lambda_2(x = 0.5) = 40$ in the above theory are plotted in Figure 5. It can be seen that the theoretically produced curves are in good agreement with the experimental values. The fitting of these curves are more pronounced at higher Dy content. Therefore, a crystalline field, acting on the RE sub-lattice, is the main origin of the observed magneto-elastic coupling, which, of course, is amplified by increasing Dy content [16,17].

In using Equation 9, relatively small higher than

second order single-ion contributions were ignored in magnetostriction (see Equation 4). At this moment, in order to consider the effect of Dy substitution on the second, fourth and sixth order of that contribution, it should be effective if the results were simulated according to the well-known $m^{l(l+1)/2}$ scaling law for magnetostriction in Equation 6. The best values of the λ_{0i} constants in Equation 6, obtained after fitting with the experimental data in Figure 5 (continuous curves are according to the lowest order magneto-elastic theory based on magnetization of 2-14-1 phase, which is obtained by choosing, $\lambda_2(x = 0.1) = 10$, $\lambda_2(x = 0.3) = 25$ and $\lambda_2(x = 0.5) = 40$ in Equation 9.) is given in Table 1. The overall agreement in each column gives support for the previous suggestion that higher-order terms in magneto-crystalline anisotropy can be more important with more Dy content in $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds.

Anisotropic magnetostriction versus external magnetic field in the temperature range of liquid nitrogen and room temperature are shown in Figure 7. The saturation behavior in the compound with $x = 0.1$ at fields above 0.7 T , is clear, while curves for $x = 0.3$ and 0.5 do not reach full saturation in the available fields. Saturation behavior at liquid nitrogen temperatures can be observed because the c -axial part of the magneto-crystalline anisotropy is near to zero ($\kappa_2^0 \rightarrow 0$) until the temperature is at about the spin re-orientation transition (Figure 3a) [10]. But, as expected from the large axial anisotropy of this compound at room temperature, it is preferred to attribute the observable saturation behavior of the anisotropic magnetostriction of the $x = 0.1$ sample at room temperature to the authors' experimental error ($\cong 4 \times 10^{-6}$). From the Dy content dependence of the spin re-orientation temperature in Figure 3a, a decreasing behavior for κ_2^0 at $T = 77 \text{ K}$ from $x = 0.1$ to $x = 0.5$ was expected. Therefore, the increase in the saturation field with Dy content once again confirms that the role of higher order anisotropy coefficients ($\kappa_4^m, \kappa_6^m, \dots$) in magnetic behavior at around $T = 200 \text{ K}$ temperatures is amplified with Dy content. This conclusion is important because it shows that higher order anisotropy coefficients must be taken into account, in addition to κ_2^0 one, in the theoretical de-

Table 1. Magneto-elastic coupling constants after fitting anisotropic magnetostriction results in Figure 5 with scaling model of Equation 6, based on RE sub-lattice magnetization of $\text{Nd}_2\text{Fe}_{14}\text{B}$ compound.

	$\lambda_{02}^* 10^6$	$\lambda_{04}^* 10^6$	$\lambda_{06}^* 10^6$
$\text{Nd}_{1.9}\text{Dy}_{0.1}\text{Fe}_{14}\text{C}$	9.70	-6.42	8.37
$\text{Nd}_{1.7}\text{Dy}_{0.3}\text{Fe}_{14}\text{C}$	21.06	-13.91	18.14
$\text{Nd}_{1.5}\text{Dy}_{0.5}\text{Fe}_{14}\text{C}$	37.39	-24.70	32.20

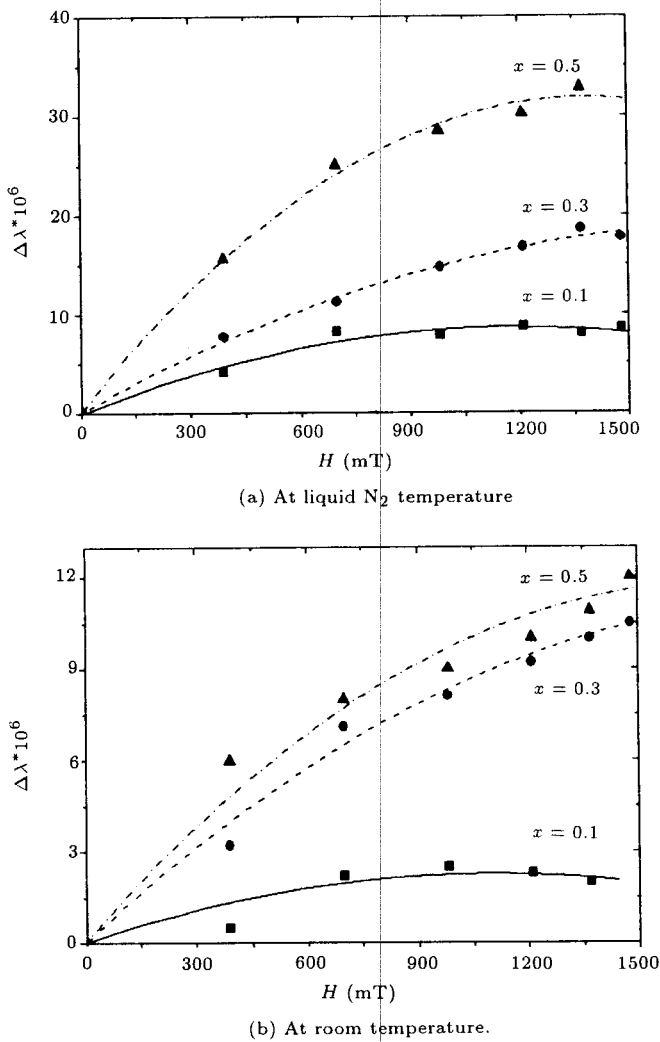


Figure 7. Variations of anisotropic magnetostriction with external magnetic field for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ samples, with $x = 0.1, 0.3$ and 0.5 .

scription of the magnetic behavior of $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds, even at temperatures more than $T \cong 220$ K.

The temperature dependence of volume magnetostriction is shown in Figure 8. It can be seen that volume magnetostriction increases rapidly at temperatures above the FOMP region. Similar behavior has been observed in the temperature dependence of the volume magnetostriction of $\text{Nd}_2\text{Fe}_{14}\text{C}$ phase by Algarabel et al. [18]. From the relative reduction of the single ion crystalline field at this temperature (Figure 5), it can be deduced that the observed increase in volume magnetostriction at $T > T_{sr}$ is originated from the two ion spin exchange coupling of RE-Fe or Fe-Fe atoms [24].

Figure 9 shows the field dependence of volume magnetostriction at room temperature for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds. A positive increase in volume magnetostriction with Dy content can be observed in this result. A kind of positive increase

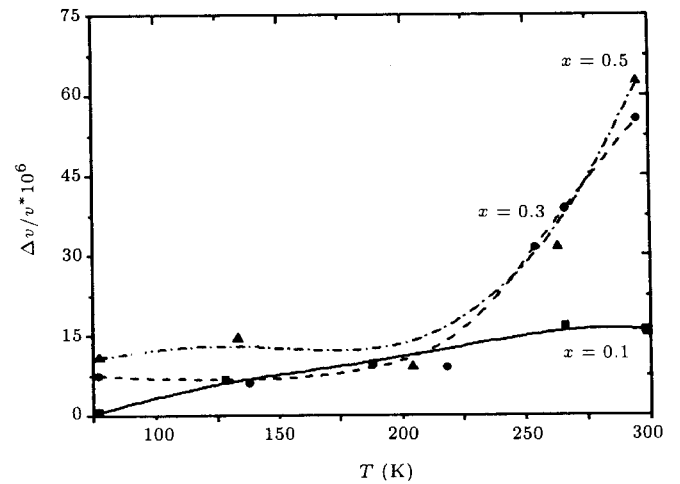


Figure 8. Volume magnetostriction at $H = 1.4$ T as a function of temperature for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ samples, with $x = 0.1, 0.3$ and 0.5 . The curves are for eye guide.

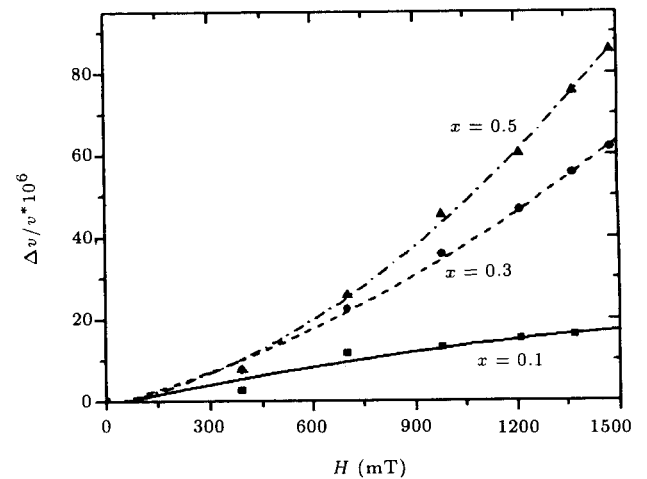


Figure 9. Volume magnetostriction as a function of external field at room temperature for $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ samples, with $x = 0.1, 0.3$ and 0.5 .

was expected based on the volume magnetostriction of $\text{Nd}_2\text{Fe}_{14}\text{C}$ and $\text{Dy}_2\text{Fe}_{14}\text{C}$ alloys from [18]. Therefore, the effect of Dy content on the room temperature isotropic part of magnetostriction ($\lambda_1^{\alpha,0}$ in Equation 8) was attributed to the positive magnetostrictive Dy-Fe anti-parallel exchange interactions. At lower temperatures, where spins of RE and Fe atoms are aligned according to a strong anisotropy, much higher magnetic fields are needed to observe large magneto-volume effects.

If one considers larger volumes of $4g$ than $4f$ sites in a tetragonal unit cell of a $\text{Nd}_2\text{Fe}_{14}\text{C}$ compound, it is expected that $4f$ sites should be preferably occupied by Dy atoms with a smaller radius than Nd. Therefore, it seems that a three sub-lattice model constructed by RE ($4f$), RE ($4g$), and Fe sub-lattices is necessary to describe the observed effect of Dy substitution on T_{SR} and T_{SP} temperatures in $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ compounds.

For some aspects of multi sub-lattice models in RE-Fe compounds see [28].

CONCLUSIONS

1. The real part of ac-magnetic susceptibility measurements show that FOMP temperature increases with Dy substitution in the $\text{Nd}_{2-x}\text{Dy}_x\text{Fe}_{14}\text{C}$ alloy, about 40 K. This is due to an increase in the higher than second order terms of the anisotropy energy of the RE sub-lattice;
2. Linear magnetostriction values become minima in the FOMP temperature region, which is due to the reduction of magnetic anisotropy in this region;
3. The temperature dependence of anisotropic magnetostriction is consistent with Callen's single ion theory. This consistency is more pronounced for samples with higher Dy content. A comparison with the scaling model indicates an increase in fourth and sixth order anisotropy parameters with Dy content;
4. The magnitude of the isotropic part of magnetostriction strongly increases with increasing Dy content after FOMP transition, which can be interpreted as a result of a positive magnetostrictive effect of anti-parallel RE-Fe exchange coupling.

ACKNOWLEDGMENT

The authors would like to express sincere thanks to Dr. H. Salamati and P. Kameli in Isfahan University of Technology for their valuable help in magnetic susceptibility measurements.

REFERENCES

1. Liu, N.C., et al. "High intrinsic coercivities in iron-rare earth-carbon-boron alloys through the carbide or borocarbide $\text{Fe}_{14}\text{R}_2\text{X}$ ($\text{X} = \text{B}_x\text{C}_{1-x}$)", *J. Appl. Phys.*, **61**, p 3574 (1987).
2. Burzo, E. "Permanent magnets based on Nd-Fe-B and Nd-Fe-C alloys", *Rep. Prog. Phys.*, **61**, p 1099 (1998).
3. de Boer, F.R., et al. "Magnetic and crystallographic properties of ternary rare earth compounds of the type $\text{R}_2\text{Fe}_{14}\text{C}$ ", *J. Magn. Mater.*, **72**, p 167 (1988).
4. Zhao, T.S., et al. "Magnetic anisotropy and crystal field in $\text{Nd}_2\text{Fe}_{14}\text{C}$ ", *J. Magn. Mater.*, **104-107**, p 1347 (1992).
5. Wohlfarth, E.P. and Buschow, K.H.J. "Ferromagnetic materials", *A Handbook on the Properties of Magnetically Ordered Substances*, **4**, North Holland Publisher (1988).
6. Pourarian, F. "Recent developments in permanent magnet production", *Iranian J. of Phys.*, **11**, p 15 (1993).
7. Marthy, A.S., Panagiotopoulos, I., Hadji, G. and Panayis, C. "Magnetic and phase transformation studies in $\text{Nd}_{15}\text{DyFe}_{75}(\text{C}, \text{B})_9$ ", *J. Appl. Phys.*, **79**, p 4616 (1996).
8. Herbest, J.F., et al. "Relationship between crystal structure and magnetic properties in $\text{Nd}_2\text{Fe}_{14}\text{B}$ ", *Phys. Rev. B*, **29**, p 4176 (1984).
9. Grossinger, R., et al. "A comparison of the magnetic anisotropy of $\text{R}_2\text{Fe}_{14}\text{B}$ with $\text{R}_2\text{Fe}_{14}\text{C}$ compound ($\text{R}=\text{Nd}, \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}, \text{Tm}, \text{Lu}$)", *J. Magn. Mater.*, **83**, p 130 (1992).
10. de Boer, F.R., et al. "Magnetic properties of $\text{Nd}_2\text{Fe}_{14}\text{C}$ and some related pseudoternary compounds", *J. Magn. Mater.*, **73**, p 263 (1988).
11. Xing, F. and Ho, W.W. "Crystallographic and magnetic behaviors of R-Fe-C compounds", *J. Appl. Phys.*, **67**, p 4604 (1990).
12. Kou, X.C. and Grossinger, R. "Spin reorientation and FOMP transition in $\text{R}_2\text{T}_{14}\text{X}$ compounds", *J. Magn. Mater.*, **95**, p 184 (1991).
13. Cebollada, F., et al. "Temperature dependence of the anisotropy constants in $\text{Nd}_2\text{Fe}_{14}\text{B}$ -based hard materials containing Dy", *IEEE Trans. on Magnetics*, **30**, p 631 (1994).
14. Kim, M.J., et al. "Spin reorientation and magnetocrystalline anisotropy of $(\text{Nd}_{1-x}\text{Dy}_x)\text{Fe}_{14}\text{B}$ ", *J. Magn. Mater.*, **224**, p 49 (2001).
15. Pareti, L., et al. "Direct observation of first-order magnetization processes in single-crystal $\text{Nd}_2\text{Fe}_{14}\text{B}$ ", *Phys. Rev. B*, **32**, p 7604 (1985).
16. Wolfers, P., et al. "Precise magnetic structures of hard ferromagnets of $\text{Nd}_2\text{Fe}_{14}\text{B}$ type as determined at low temperature from single crystal neutron diffraction", *Proceeding of "MSM99" Conf.*, Tehran-Iran, World Scientific, p 991 (2000).
17. Pourarian, F. "Development of high performance permanent magnets based on Nd-Fe-B system", *Proceeding of "MSM99" Conf.*, Tehran-Iran, World Scientific, p 975 (2000).
18. Algarabel, P.A., et al. "High field magnetostriction and magnetic thermal expansion of $\text{Re}_2\text{Fe}_{14}\text{B}$ hard intermetallics", *J. Magn. Mater.*, **114**, p 161 (1992).
19. Buschow, K.H.J., *Handbook of Ferromagnetic Materials*, **7**, Ch 5, North Holland Publisher (1988).
20. Callen, E.R. and Callen, H.B. "Static magnetoelastic coupling in cubic crystals", *Phys. Rev.*, **129**, p 578 (1963).
21. Callen, E.R. and Callen, H.B. "Magnetostriction, forced magnetostriction and anomalous thermal expansion in ferromagnets", *Phys. Rev. A*, **139**, p 455 (1965).
22. Kuz'min, M.D. "Magnetostriction of DyFe_2 and HoFe_2 : Validity of single-ion model", *J. Appl. Phys.*, **89**, p 5592 (2001).

23. de Mooij, D.B. and Buschow, K.H.J. "Formation and magnetic properties of the compounds $R_2Fe_{14}C$ ", *J. Less-Common Met.*, **142**, p 349 (1988).
24. Pourarian, F. and Tajabor, N. "Magnetostriction in some rare-earth- Co_5 compounds", *Phys. Stat. Sol.*, **61**, p 637 (1980).
25. Lee, E.W. and Pourarian, F. "Magnetoelastic properties of rare earth compounds II", *Phys. Stat. Sol. (a)*, **34**, p 383 (1976).
26. Lachaisserie, E.d.T.d., *Magnetostriction 'Theory and Applications'*, CRC Press, Boca Raton (1992).
27. Onodera, H., et al. "Mossbauer study of the intermetallic compound $Nd_2Fe_{14}B$ ", *J. Magn. Magn. Mater.*, **68**, p 6 (1987).
28. Yu, M. and Zhang, A. "Second-order spin reorientation reform in the two-sublattice ferromagnetic system", *J. Magn. Magn. Mater.*, **232**, p 60 (2001).