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Thermal expansion anomaly and magnetostriction of $Nd_2Fe_{14}Si_3$ intermetallic compound

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ABSTRACT

The thermal expansion and magnetostriction of $Nd_2Fe_{14}Si_3$ polycrystalline intermetallic compound was measured, using the strain gauge method in the temperature range 77–590 K under applied magnetic fields up to 1.5 T. Results show that both linear thermal expansion and linear thermal expansion coefficient exhibit anomalies below Curie temperature. A pronounced positive spontaneous volume magnetostriction has been observed below the Curie temperature. The spontaneous volume magnetostriction decreases with Si substitution in Nd_2Fe_{17} compound. The saturation trend of the anisotropic magnetostriction below 115 K depends on the saturation of the λ^{γ} mode. In the low field region ($\mu_0 H < 0.3$ T), a minimum appears in the isothermal curves of the volume magnetostriction that may be attributed to the domain walls motion and rotation of the magnetic moments of domains toward the applied field direction. The results of the spontaneous and anisotropic magnetostriction are discussed based on the local magnetic moment model and irreducible magnetoelastic coupling modes, respectively.

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1. Introduction

Intermetallic compounds based on rare-earth (R) and transition metal (T) elements have an important role from technological applications and fundamental viewpoints [1]. The large magnetic anisotropy originates from R sublattice while high Curie temperature (T_C) and spontaneous magnetization (M_S) are attributed to T sublattice in these material series [2]. The binary compounds R_2Fe_{17} are one of widest group of R–T intermetallics. These compounds are either ferromagnetic system with R = light rare-earth or ferrimagnetic system with R = heavy rare-earth elements [3]. The compounds crystallize either in the rhombohedral structure of Th_2Zn_{17}-type (for light R) or hexagonal structure of Th_2Ni_{17}-type (for heavy R), where Fe atoms occupy four non-equivalent sites and R ions are located on one or two different crystallographic sites, respectively [4]. Previous results show that the Curie temperature of R_2Fe_{17} is between 225 to 480 K, which is far less than for the bcc iron (T_C = 1073 K) [3]. The value of T_C is due to Fe–Fe exchange interaction which is strongly dependent on the d_{Fe-Fe} (Fe–Fe interatomic distance). The critical bond length between a positive exchange interaction (ferromagnetic) and a negative exchange interaction (antiferromagnetic) is about 2.45 Å [5]. Therefore, the competition of ferromagnetic ($d_{Fe-Fe} > 2.45$ Å) and antiferromagnetic ($d_{Fe-Fe} < 2.45$ Å) interactions of Fe–Fe pairs originates the low T_C in these compounds. Thus, for increasing T_C, d_{Fe-Fe} must be optimized with the change of lattice parameters. The low Curie temperature of R_2Fe_{17} compounds restricts their practical applications as permanent magnet and high temperature magnetostrictive devices. Hence, many investigations were performed on R_2Fe_{17} with the aim of improving their T_C. For example, partial substitution of Si, Cr, Ga and Al for Fe atoms in several R_2Fe_{17} compounds changes lattice parameters and leads to significant increase in T_C and to modifying their magnetic anisotropy [6–11]. The anomalous thermal expansion and spontaneous magnetostriction is another result from this competition [12]. In the past two decade, many articles on the magnetostriction of R_2Fe_{17} single crystal and polycrystalline compounds were reported [13–17]. However, effects of Si substitution on magnetoelastic properties such as thermal expansion and magnetostriction have been less considered until now [18–20]. Following our investigation on magnetoelastic properties of $R_2Fe_{17-x}Si_x$ (R = Y, Er and Tm) compounds [21], we report the thermal expansion and magnetostriction of $R_2Fe_{14}Si_3$ (R = Ce and Nd) and then estimate the contribution of Nd and Fe sublattice to magnetoelastic properties.

2. Experimental details

The $R_2Fe_{14}Si_3$ (R = Ce and Nd) polycrystalline samples were prepared by induction melting of the constituent materials of 99.9% purity under purified Ar atmosphere. As-cast ingots were heat-treated at 900 °C for 2 days and then rapidly

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cooled to room temperature. Powder X-ray diffraction (with Cu Kα radiation) patterns of the annealed samples was used to determine phase purity, crystal structure and lattice parameters. X-ray diffraction of $R_2Fe_{14}Si_3$ compounds indicate that the prepared samples to be almost single phase that crystallize in rhombohedral structure of Th_2Zn_{17}-type, however, a few percent of alpha iron were observed as a second phase in $Nd_2Fe_{14}Si_3$ sample. The presence of such impurity phase has also been reported in earlier publications [22,23]. The lattice parameters are found to be $a = 8.443$ Å, $c = 12.434$ Å and $a = 8.512$ Å, $c = 12.464$ Å for R = Ce and Nd, respectively. These results are consistent with the literature [8].

Thermal expansion and magnetostriction measurements were performed by using a strain gauge method. Disk-shaped samples (10 mm diameter, 3 mm thick) were used, which cut from the annealed bulk ingots. Magnetic fields were applied up to 1.5 T, with temperature ranging from 77 to 590 K. The linear thermal expansions $\Delta l/l = [l(T) - l(77\,\mathrm{K})]/l(77\,\mathrm{K})$ were deduced by measuring the relative change of length of the samples versus temperature. The magnetostriction were measured (with an accuracy of 2×10^{-6}) parallel (longitudinal magnetostriction, λ_l) and perpendicular (transverse magnetostriction λ_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 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

The linear thermal expansion curves, $\Delta l/l(T)$ and linear thermal expansion coefficient, $\alpha(T) = \frac{d}{dT}[\Delta l/l(T)]$ of $R_2Fe_{14}Si_3$ (R = Ce and Nd) compounds are shown in Fig. 1. It can be seen that both $\Delta l/l(T)$ and $\alpha(T)$ of samples exhibit an anomalous behaviour near T_C. Beyond T_C in the paramagnetic region, $\Delta l/l(T)$ behaves linearly. The spontaneous linear magnetostriction, $[\Delta l/l(T)]_m$, was determined by subtracting the lattice thermal expansion, $[\Delta l/l(T)]_{\mathrm{latt}}$, from the total experimental linear thermal expansion, $[\Delta l/l(T)]_{\mathrm{exp}}$ i.e. $[\Delta l/l(T)]_m = [\Delta l/l(T)]_{\mathrm{exp}} - [\Delta l/l(T)]_{\mathrm{latt}}$. 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, $T_D = 450$ K [18,20], and Grüneisen relation, $\alpha(T) = \kappa\gamma C_V/3$ where α is the phonon anharmonic thermal expansion coefficient, κ the isothermal compressibility, γ the Grüneisen parameter, and C_V is the specific heat. Assuming $\Delta l/l(T)$ is isotropic for polycrystalline sample; 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 $R_2Fe_{14}Si_3$ (R = Ce and Nd) compounds. The results show the decrease of ω_s from 5.7×10^{-3} and 7.0×10^{-3} to about zero at T_C for Ce and Nd containing compounds, respectively. The comparison of these values with those of Ce_2Fe_{17} and Nd_2Fe_{17} indicate that ω_s decrease with Si substitution [12,13]. The nonzero remaining values of ω_s above T_C reflect the existence of a short-range magnetic ordering in the paramagnetic state, similar to other 2–17 compounds [12,18,20].

In order to investigate the spontaneous magnetostriction of these compounds and the effect of Si substitution on $\omega_s(T)$ of R_2Fe_{17}-type, we use the local magnetic moment model (the Heisenberg model). In this model, ω_s is proportional to two-ion isotropic spin correlation functions $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ [24], which is called as the magnetic exchange interaction between i and j lattice sites. It is well known that in R-Fe compounds, there are three exchange interactions included J_{FeFe} between the Fe spins, J_{RFe} between the rare earth spin and Fe spins, and J_{RR} between rare earth spins. J_{FeFe} is dominant in comparison to J_{RFe}, whereas J_{RR} contribution is small and can be neglected [22]. Therefore, J_{FeFe} has the major contribution to ω_s in these compounds. The total Fe-Fe exchange interaction is not only determined by the exchange parameter J^{ij}_{FeFe} (exchange integral between Fe{i} and Fe{j}, where {i} and {j} are Fe atoms for equivalent and non-equivalent sites) but also by Fe magnetic moment and the number of iron nearest neighbors [20,25]. The signs and magnitudes of exchange integral as a function of the distance of the Fe-Fe pairs determine using famous Bethe-Slater curve (Fig. 3). It is clear that with an increase in the distance of the Fe–Fe pairs, the exchange integrals modify from negative to positive. When the distance is smaller than 2.45 Å, the exchange interactions between the Fe atoms are negative; while the Fe atoms are located at a larger distance the interactions are positive. Moreover, the positive interactions increase at first and then decrease. Previous results showed that when Si atoms replace Fe in $R_2Fe_{17-x}Si_x$ (R = Ce and Nd) alloys, Si atoms avoid the 6c site, substitute preferentially into the 18 h site, fill the 9d site steadily and fill the 18f site only at high Si content [26–28]. Therefore, the magnetic moments, the bond lengths and the effective number

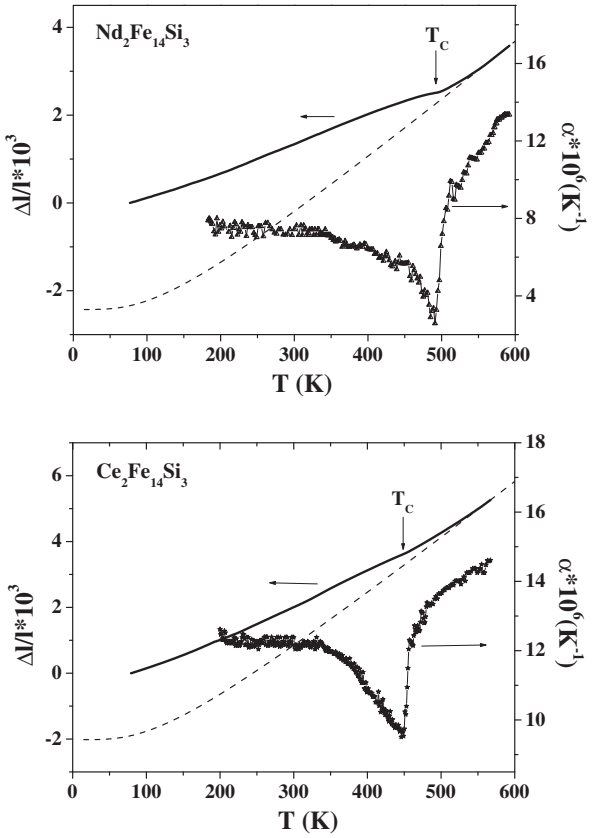

Fig. 1. Linear thermal expansion $\Delta l/l(T)$ and linear thermal expansion coefficient $\alpha(T)$ for $R_2Fe_{14}Si_3$ (R = Ce and Nd) compounds, The dashed lines represent the calculated phonon contribution to the thermal expansion as discussed in the text. The arrows indicate the Curie temperatures.

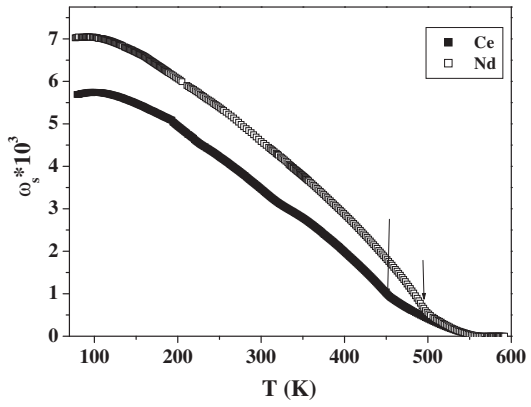

Fig. 2. Temperature dependence of the spontaneous volume magnetostriction (ω_s) for $R_2Fe_{14}Si_3$ (R = Ce and Nd) compounds. The arrows indicate the Curie temperatures.

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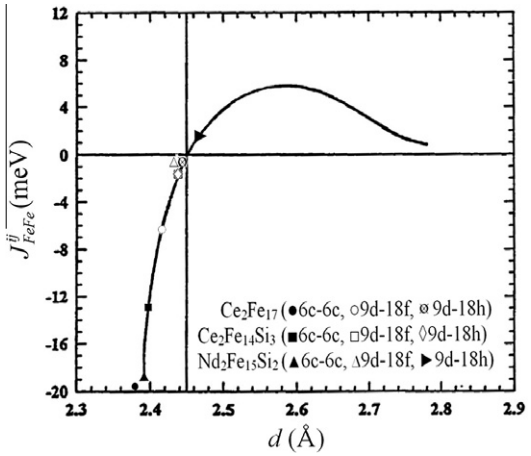

Fig. 3. The exchange integrals, J^{ij}_{FeFe} as a function of the distances between the Fe-Fe pairs. The bond lengths of 6c–6c, 9d–18f, and 9d–18h sites for Ce_2Fe_{17} (circles), $Ce_2Fe_{14}Si_3$ (squares) and $Nd_2Fe_{15}Si_2$ (triangles) compounds [21,25].

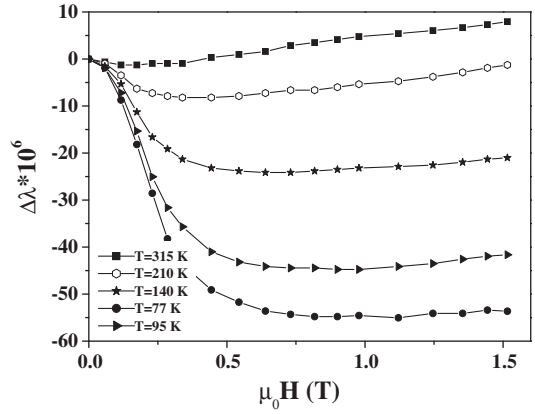

Fig. 4. Field dependence of the anisotropic magnetostriction of $Nd_2Fe_{14}Si_3$ compound at different temperatures. In this and all following figures, the lines are guides for the eyes.

of iron nearest neighbors change by Si substitution. The three sets of bond lengths for Ce_2Fe_{17}, $Ce_2Fe_{14}Si_3$ and $Nd_2Fe_{15}Si_2$ are depicted on the Bethe-Slater curve in Fig. 3, using the Neutron diffraction measurements on these compounds [22,26–28]. The anomalous thermal expansion and Invar effect in R_2Fe_{17}-type below room temperature is due to the negative magnetic interaction between the 6c–6c, 9d–18f, and 9d–18h sites [5]. Here, we calculate approximately J_{FeFe} (Heisenberg Hamiltonian) for these sites as:

$$\begin{aligned} J_{FeFe} &= \sum_{i,j=c-c,d-f,d-h} N_{i-j} m_i m_j J^{ij}_{FeFe} \\ &= N_{c-c} m_c m_c J^{c-c}_{FeFe} + N_{d-f} m_d m_f J^{d-f}_{FeFe} + N_{d-h} m_d m_h J^{d-h}_{FeFe} \\ &= 1 \cdot (2.5)^2 \cdot (-17.5) + 4 \cdot (1.7) \cdot (2.4) \cdot (-1.5) \\ &\quad + 4 \cdot (1.7) \cdot (1.7) \cdot (+2.5) = -83.1 \text{ meV} \end{aligned} \quad (1)$$

$$\begin{aligned} J_{FeFe} &= 1 \cdot (2.1)^2 \cdot (-18) + (N_{d-f} \leqslant 4) \cdot (1.7) \cdot (2.4) \cdot (-1.5) \\ &\quad + (N_{d-h} \leqslant 4) \cdot (1.7) \cdot (2.4) \cdot (2.5) \leqslant -62.7 \text{ meV} \end{aligned}$$

for Nd_2Fe_{17} and $Nd_2Fe_{14}Si_3$ compounds, respectively [27,28]. In Eq. (1), N_{i-j}, m_i and m_j are the effective number of bonds between i and j sites, the magnetic moment of i and j sites, respectively. For simplification, the 6c, 9d, 18f, 18h sites are labeled with c, d, f, and h. It is noticed that in above calculation we use the bond lengths of $Nd_2Fe_{15}Si_2$ for $Nd_2Fe_{14}Si_3$ sample. Presence of ($\leqslant$) in second term indicate that the effective number of d–f and d–h bonds in $Nd_2Fe_{14}Si_3$ is lower than that of Nd_2Fe_{17} because of Si substitution in these sites. As a result of the above calculation, the decreasing of $\omega_s(T)$ and the disappearing of the Invar effect of $Nd_2Fe_{14}Si_3$ compared to Nd_2Fe_{17} can be attributed to the increasing of the exchange interaction from −83.1 meV to J_{FeFe} = −62.7 meV [12,22,26–28]. Such behavior has been also observed in hexagonal structure of Th_2Ni_{17}-type [18,20]. On the other hand, the comparison of spontaneous magnetostriction for Ce and Nd containing compounds in Fig. 2 show that $\omega_s(Nd) > \omega_s(Ce)$. This result is also a consequence of $J_{FeFe}(Nd) < J_{FeFe}(Ce)$ based on this model. Moreover, the comparison of ω_s for Ce and Nd containing compounds indicate that the major contribution to ω_s originates from ω_s of the Fe sublattice. Similar results have also been observed in other related intermetallic compounds [29,30]. Finally, the decrease of ω_s with temperature can be attributed to the decrease of the magnetic moment and the magnetic exchange interaction values with temperature increasing.

Fig. 4 shows the anisotropic magnetostriction ($\Delta\lambda$) of $Nd_2Fe_{14}Si_3$ compound as a function of applied field at some selected temperatures. It can be seen that the $\Delta\lambda$ curves exhibit different behaviors which strongly depend on temperature. At low temperature ($T < 115$ K), the $\Delta\lambda$ curves reach complete saturation in $\mu_0 H < 0.8$ T. In the temperature range 115 K < T < 210 K, $\Delta\lambda$ strongly decreases at a low field, reach a minimum in low-field region, and then monotonically increases. With more increasing temperature, $\Delta\lambda$ reveres its sign for the high applied fields. Also, the values of applied threshold field in inflection point on isothermal curves of $\Delta\lambda$ decrease with increasing temperature. The temperature dependence of $\Delta\lambda$ for this compound at selected applied field is shown in Fig. 4. As one can see the anisotropic magnetostriction increases with temperature. Finally, magnetostriction compensation occurs at a point (T_0) which is different at typical applied fields. A Similar magnetostriction compensation point has been also observed in Nd_2Fe_{17} compound [16]. Further, comparison of $\Delta\lambda$ values for $Nd_2Fe_{14}Si_3$ and Nd_2Fe_{17} in ref. [16] shows that $\Delta\lambda$ decreases by Si substitution in low temperature.

For detail analysis of the magnetostriction results of our polycrystalline samples, we use the irreducible magnetoelastic coupling modes (i.m.c.m) in the notation of Callen [16,24]. For polycrystalline materials with axial symmetry (such as the studied compounds), by averaging over all the space angles the anisotropic magnetostriction $(\Delta\lambda)_{ava}$ can be explained versus i.m.c.m as [16,24],

$$(\Delta\lambda)_{ava} = \frac{2}{15}(\sqrt{3}\lambda^{\alpha}_{22} + 3\lambda^{\gamma} + 2\lambda^{\varepsilon}) + \text{higher order terms} \quad (2)$$

where, λ^{α}_{22} mode represents variations of c/a ratio, λ^{γ} refers to the breaking of cylindrical symmetry in the basal plane and λ^{ε} a shear in a plane containing the c-axis [14,15]. This means that the anisotropic magnetostriction in polycrystalline materials is attributed to the single-ion crystal electric field (CEF) and no exchange striction terms (λ^{α}_{11} *and* λ^{α}_{12}) contribute to $\Delta\lambda$. The Eq. (2), using the measurements on the same single crystal and polycrystalline compounds such as Y_2Fe_{17} [14,17] and Ho_2Fe_{17} [15,17] has been approved. Our compound is a polycrystalline sample with large magnetocrystalline anisotropy (H_A) of an easy plane type with the b-axis as the easy magnetization direction as reported in [11]. The magnitude of anisotropy field below 115 K in this compound is H_A > 14 T which is higher than our available applied fields. Thus, $\Delta\lambda$ depends on rotation of magnetic moments in basal planes by applied field up to 1.5 T. Based on previous reports on single crystalline compounds with basal plane anisotropy; it was observed that λ^{ε} mode is negligible with respect to other modes. Also, λ^{α}_{22} is found to be small at low magnetic fields and low temperatures [15]. Moreover, λ^{γ} mode saturates in low fields and its value is independent of the applied fields [14,15]. Therefore, the saturation trend below 115 K may be attributed to the saturation of the λ^{γ} mode. The magnetisation

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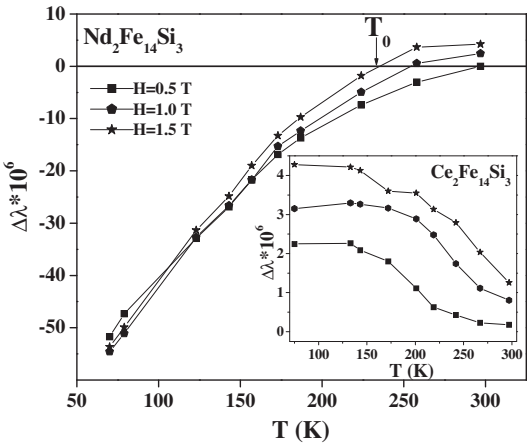

Fig. 5. Temperature dependence of the anisotropic magnetostriction of $Nd_2Fe_{14}Si_3$ compound at the selected magnetic fields. The arrows refer to the compensation point (T_0). The inset shows the temperature dependence of $\Delta\lambda$ in $Ce_2Fe_{14}Si_3$ sample at the selected magnetic fields.

information showed that the anisotropy field within basal plane decreases with increasing temperature [11]. Consequently, λ^{γ} mode decreases with temperature. On the other hand, in single crystalline compound is observed that λ_{22}^{α} mode increase with temperature [14–15]. Therefore, gradually λ_{22}^{α} mode is enhanced in $Nd_2Fe_{14}Si_3$ sample. Hence, the saturation behaviour is disappeared and then inflection point is exhibited in $\Delta\lambda$ curves above 115 K. the first, inflection points are exhibited in medium field region but with more increasing temperature, λ_{22}^{α}mode dominates λ^{γ} mode in low fields so that inflection points take place in the low field region. Finally, the occurrence of magnetostriction compensation point in Fig. 5 show that λ^{γ} is negative and λ_{22}^{α} has positive value. In order to determine Nd and Fe sublattice contribution to magnetostriction, we measure the magnetostriction of $Ce_2Fe_{14}Si_3$ compound (the inset of Fig. 4) which has the same rhombohedral structure and planar magnetic anisotropy such as $Nd_2Fe_{14}Si_3$ [11,31]. Since, Ce ion is tetravalent and has no 4f-derived magnetic moment; therefore, it can be assumed that has negligible contribution to the magnetostriction. Hence, the magnetostriction of Fe sublattice is obtained from $Ce_2Fe_{14}Si_3$ magnetostriction. Results show that Fe sublattice contribution to magnetostriction is positive and one order of magnitude lower than that of Nd sublattice. Similar results have also been observed in the magnetostriction of other related intermetallic compounds [32].

The volume magnetostriction ($\Delta V/V$) of the $Nd_2Fe_{14}Si_3$ compound is plotted versus applied field at selected temperature in Fig. 6. It is clear that the volume magnetostriction curves in high field region are positive and increase with the applied fields. However, in the low field region ($\mu_0 H < 0.3$ T), a minimum appears in the isothermal curves of $\Delta V/V$. By decreasing temperature these minimum not only become deeper but are also displaced at lower applied fields. This effect may be attributed to the domain walls motion and rotation of the magnetic moments of domains toward the applied field direction that are accompanied by negative volume magnetostriction.

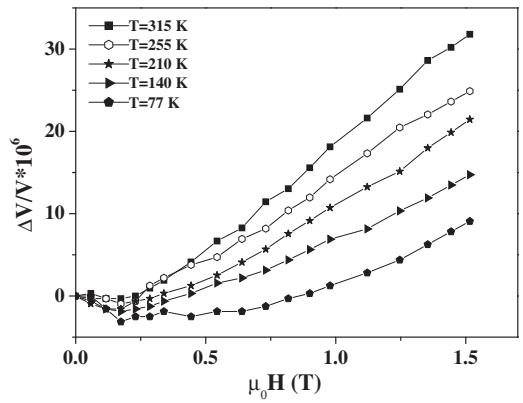

Fig. 6. Field dependences of the volume magnetostriction of $Nd_2Fe_{14}Si_3$ compound at some typical temperatures.

4. Conclusions

We investigated the magnetoelastic properties of $R_2Fe_{14}Si_3$ (R = Ce and Nd) alloys. The spontaneous volume magnetostriction decreases and the Invar effect disappears with Si substitution that can be ascribed to increasing of exchange interaction between 6c–6c, 9d–18f, and 9d-18 h sites. The appearance of the magnetostriction compensation point in $\Delta\lambda$ curves of $Nd_2Fe_{14}Si_3$ show that λ^{γ} is negative and λ_{22}^{α} has positive sign. The measurements of the magnetostriction on $Ce_2Fe_{14}Si_3$ and $Nd_2Fe_{14}Si_3$ samples exhibit that Fe sublattice contribution to magnetostriction is positive and one order of magnitude lower than that of Nd sublattice. The volume magnetostriction curves in high field region are positive and increase with the applied fields. However, in the low field region ($\mu_0 H < 0.3$ T), a minimum appears in the isothermal curves of $\Delta V/V$.

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