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Study of the magnetostrictive strains in $Pr_6Fe_{11}Ga_3$ alloy

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Abstract

Magnetoelastic properties of the $Pr_6Fe_{11}Ga_3$ alloy are studied by magnetostriction and thermal expansion measurements. The effects of short- and long-range magnetic ordering processes about Curie temperature clearly appear in the temperature dependence of the spontaneous magnetostriction as two increasing steps with decreasing temperatures. Thermal variations of the total magnetocrystalline anisotropy introduce pronounce changes in the isofield curves of the forced magnetostriction as a negative minimum below 200 K, a compensation phenomena about 250 K, and a positive maximum between 250 K and $T_c = 320$ K. The observed behavior of magnetostriction is discussed in terms of the competitive anisotropies of Pr and Fe sublattices and coupling magnetostrictive constants.

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Keywords: Magnetostriction; Thermal expansion; Magnetocrystalline anisotropy

1. Introduction

$RE_6Fe_{14-x}M_x$ intermetallic alloys are recognized as coercivity enhancer and were found as a secondary phase in the grain boundary of $RE_2Fe_{14}B$ permanent magnets [1,2]. They crystallize in the tetragonal I4/mcm structure in which RE atoms mainly surrounded by other RE atoms [3,4]. There is a great debate about magnetic interactions in these alloys. This originates firstly from their complex crystal structure, and secondly, from the tiny traces of magnetic impurities, such as RE_2Fe_{17} ferromagnetic phase, that usually appears in these alloys but could not be traced by X-ray and neutron diffractometries [5]. Magnetic measurements and MÖssbauer spectroscopy studies show that there are three interacting noncollinear sublattices, formed by RE (16*l*), RE (8*f*) and Fe atoms. These sublattices should be considered for describing the magnetic properties of these alloys [6,7].

The good solubility of Al and Ga elements in $RE_6Fe_{14-x}[Ga (Al)]_x$ makes these compounds to be attractive for studies of their physical properties. Among these intermetallics, the single-phase alloy of $RE_6Fe_{11}Ga_3$ with RE = Pr or Nd, can easily be formed [7]. Previous results on the Pr-based alloys of these families show an antiferromagnetic spin structure, which is believed to be originated from the competitive magnetocrystalline anisotropies of the RE (16*l*), RE (8*f*) and Fe sublattices [8]. Magnetic behavior of $Pr_6Fe_{14-x}Ga_x$ is found to be similar to $Pr_6Fe_{14-x}Al_x$, which is attributed to the close values of the electronegativity and atomic radius of Ga and Al. Although Pr atoms at 16*l* crystallographic site exhibit large uniaxial anisotropy [6], however, the exchange interaction between Fe($16l_2$) and Pr(8*f*) promotes easy plane ordering of the Pr(8*f*) atoms so that easy plane magnetization is retained over 1.5–400 K interval. This creates frustration of Pr(16*l*) ordering, which is weakened as magnetization of Pr(16*l*) sublattice increases with decreasing temperature, specially below 100 K.

Due to large magnetocrystalline anisotropy of Pr atoms, the experimental magnetization of $Pr_6Fe_{11}Ga_3$ do not saturate in the presence of applied fields up to 7 T. The compound exhibits some type of metamagnetic first-order magnetic processes (FOMP) even at room temperature [6].

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Magnetization measurements indicate that, the long-range order of ferrimagnetic phase reduces to the short-range ferromagnetic order as temperature approaches to the Curie temperature ($T_c \cong 320$ K) [7]. These two magnetically ordered phases are separated by magnetization compensation phenomena.

Up to now, very little is known about magnetoelastic interactions in $Pr_6Fe_{11}Ga_3$ alloy. However, it is interesting to study the magnetostrictive properties and magnetoelastic anisotropy of $RE_2Fe_{14}BGa$ magnets [9]. In this work, attempt is made to investigate magnetostriction and thermal expansion of this alloy.

2. Experimental

The polycrystalline $Pr_6Fe_{11}Ga_3$ alloy was prepared by RF induction melting of the constituent elements, Pr (99.95%), Fe and Ga (99.99%). This alloy wrapped in a tantalum foil, encapsulated in a quartz tube under argon atmosphere, and annealed at ~600 °C for 8 days to obtain single-phase material. The microstructure and phase purity of the sample are studied using scanning electron microscopy and X-ray diffraction analysis. These studies show that the sample contained grains of $Pr_6Fe_{11}Ga_3$ phase, with about 5 μm size.

Magnetostrictive strains were measured using familiar strain gage method in the presence of external fields up to 15 kOe and in temperature range from 77 to 300 K. These measurements were performed on the disk-shaped samples (diameter = 6 mm, thickness = 2 mm) as external field applied parallel (λ_l) and perpendicular (λ_t) to the gage length. Then, anisotropic and volume magnetostrictions are calculated using $\Delta\lambda = \lambda_l - \lambda_t$ and $\omega = \lambda_l + 2\lambda_t$ relations, respectively. For seeking reproducibility of the results, all measurements were repeated by iterative warming and cooling of the sample. Also, no hysteric behavior and no significant difference were observed between magnetostriction and thermal expansion of samples that have been cut along three perpendicular directions of the bulk alloy. This implies absence of thermal cracking and any preferential magnetic orientation in the $Pr_6Fe_{11}Ga_3$ annealed alloy. The thermal expansion is deduced by measuring relative change of the length at each temperature, $\Delta l/l_{N_2} = (l_{(T)} - l_{N_2})/l_{N_2}$, then the thermal expansion coefficient (α) was calculated based on the linear approximation, $\Delta l/l_{N_2} = \alpha\Delta T$.

3. Results and discussion

Fig. 1 shows the thermal expansion of the studied alloy between 78 and 350 K. These results show metallic behavior below 180 K and some anomalies at higher temperatures (see α curve in Fig. 2). It is known that the thermal expansion of the magnetic materials is usually influenced by the magnetization in addition to the lattice vibrations. The magnetic contribution can be estimated by extrapolation of the linear paramagnetic behavior that is observed above $T_c = 320$ K to the ferromagnetic region ($T < T_c$). The difference between experimental curve and extrapolated line is called spontaneous magnetostriction, λ_{ex}, that refers to the change of the crystallographic unit cell volume with temperature. The temperature dependence of the qualitative values of λ_{ex} is shown in Fig. 2. As temperature decreases below $T = 335$ K, λ_{ex} increases in a sequence of two steps. First step is located in the vicinity of Curie temperature ($T_c = 320$ K) and the second step starts at about 295 K. The small positive λ_{ex} that is appeared in the first step can be attributed to magnetoelastic effects of the short-range ferromagnetic ordering process. In the $RE_6Fe_{14-x}M_x$ family of alloys, the short-range magnetic order may originate from the Fe–Fe exchange interaction. These magnetic correlations usually are considered as the primary step for expanding a macroscopically ordered magnetic phase over the volume of magnetic materials [10]. Considering the crystal and magnetic structure of the studied alloy in Fig. 3, the preliminary short-range

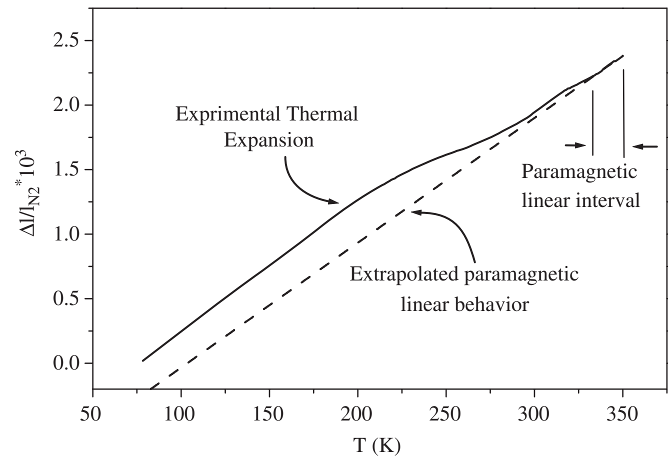

Fig. 1. Experimental thermal expansion curve of $Pr_6Fe_{11}Ga_3$ alloy between 78 and 350 K. The dash line exhibits extrapolation of the paramagnetic behavior to below Curie temperature ($T_c = 320$ K).

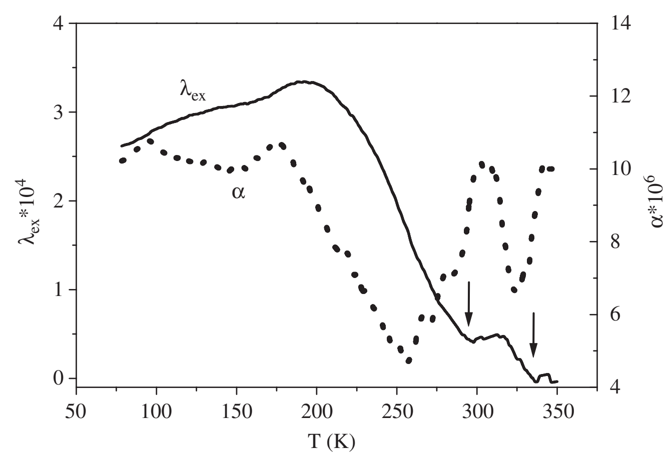

Fig. 2. Temperature dependence of the qualitative spontaneous magnetostriction and thermal expansion coefficient of $Pr_6Fe_{11}Ga_3$ alloy. Arrows indicate to the beginning of the short- and long-range magnetic ordering processes with decreasing temperatures.

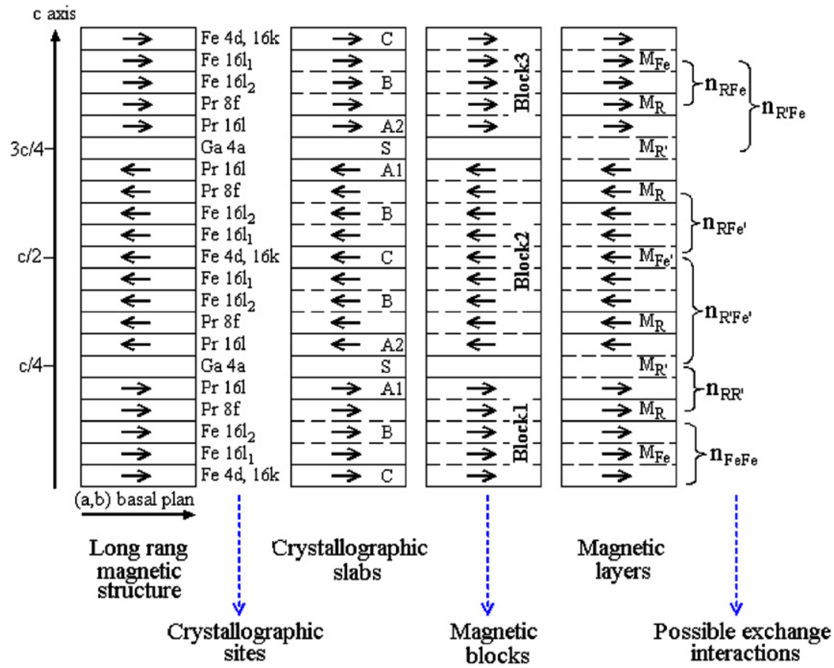

Fig. 3. the conventional concepts in description of the crystal and magnetic structure of $Pr_6Fe_{11}Ga_3$ alloy based on the neutron diffraction [5,7]. Estimation of the exchange field parameters shows that $|n_{R'Fe}|$ $(|n_{R'Fe'}|)<|n_{RR'}|<|n_{FeFe'}|\ll|n_{RFe}|<|n_{FeFe}|$, and $n_{R'Fe}\approx n_{R'R}\approx 0$. Consequently, the R(8$f$) moments are strongly and ferromagnetically coupled to the Fe moments while R′(16l) ones behave paramagnetically or become ordered at a temperature well below $T_N\approx 250$ K.

magnetic orders can be originated from n_{Fe-Fe} intralayer ferromagnetic exchange interaction.

The second step of the λ_{ex} starts at about 295 K, with decreasing temperatures. Here, the increase of λ_{ex} is greater than that of the first step. This positive λ_{ex} can be attributed to the gradual occurrence of the long-range ordering of the magnetic moments that is concurrent with the low-temperature ferrimagnetic phase of $Pr_6Fe_{11}Ga_3$. As temperature decreases down to 190 K the magnitude of λ_{ex} reaches its maximum value (about $+3.3\times10^{-4}$). In $Pr_6Fe_{11}Ga_3$ alloy, it is believed that the exchange coupling of Fe–Fe atoms are completed around 290 K and their alignment is restricted by the anisotropy of Pr sublattices at lower temperatures [5–7]. In fact, the long-range order starts by activation of the Fe(16l_2)–Pr(8f) interlayer planar ferromagnetic interaction (n_{RFe}), and will be completed by the antiferromagnetic arrangement of blocks separated by the nonmagnetic S slabs (see Fig. 3). As discussed above, Fe (16l_2)–Pr(8f) exchange interactions promote basal anisotropy of the Pr(8f) atoms over inherently larger axial anisotropy of Pr(16l) [5]. Below 190 K, the observed small decrease of λ_{ex} with decreasing temperature (Fig. 2) may originate from higher order terms in the magneto-crystalline anisotropy energy which is accompanied with the gradual ordering of the frustrated Pr(16l) moments. Considering that the basal antiferromagnetic order is conserved over the entire temperature range of magnetic ordering, we do not expect additional jump in the λ_{ex} curve.

Fig. 4 shows the anisotropic magnetostriction as a function of applied field for typical temperatures. It is clear that below an applied threshold field ($\mu_0H<0.4$ T at 78 K,

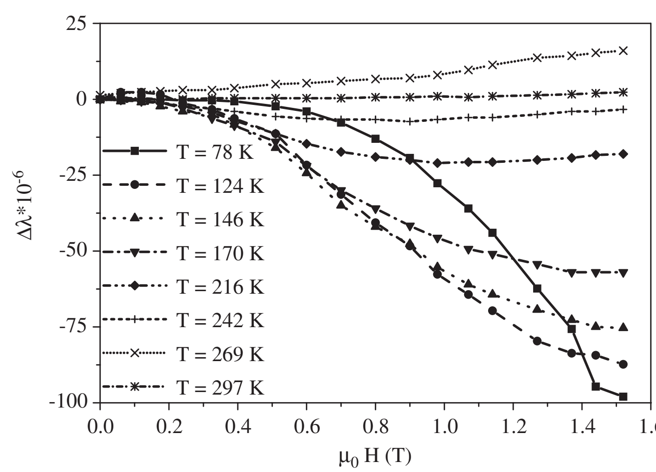

Fig. 4. Isothermal curves of the anisotropic magnetostriction of $Pr_6Fe_{11}Ga_3$ alloy as a function of applied field at selected temperatures.

and, less than 1 T at 216 K), magnetostrictive strains are small and smoothly increase with increasing fields. Also, the threshold field increases with decreasing temperatures, especially below 128 K. Considering that the induced magnetization at this region of fields mainly originates from the extending domains, thus the result confirms existence of the strong pinning centers in the alloy that prevents easy movement of the domain walls. In fact, the localized 4f orbital of the RE atoms (here, Pr) creates large magnetocrystalline anisotropy that is responsible for providing strong pining centers in the RE–TM intermetallics [10]. Hence, the observed increase of the H_a with decreasing temperatures may be attributed to the natural

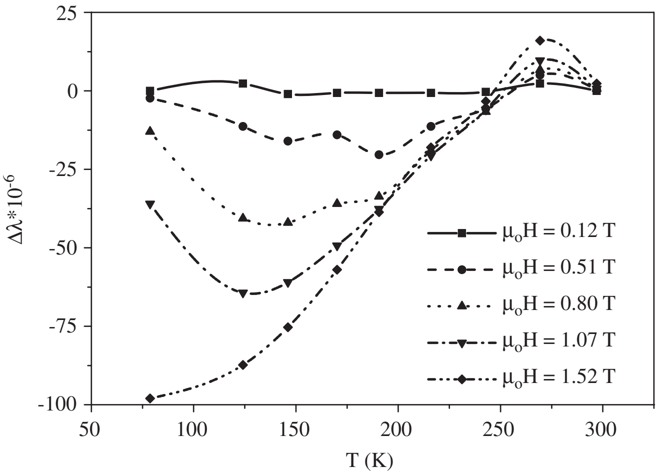

Fig. 5. Temperature dependence of the anisotropic magnetostriction of $Pr_6Fe_{11}Ga_3$ alloy at selected applied fields.

enhancement of the magnetocrystalline anisotropy of Pr sublattices. This behavior was also confirmed using neutron thermodiffractometry method [5]. Although pining of the domain walls usually is accompanied by the hysteresis in the magnetization of RE–TM compounds, but no hysteresis was observed in the $\Delta\lambda$ measurements. This observation is consistent with the negligible magnetostrictive strains of the domain wall stretching observed below the threshold fields in Fig. 4.

As applied field increases above the threshold field, the magnetic moments gradually rotate toward the field direction in addition to the domain extension. This induced magnetization is accompanied with the presence of a negative magnetoelastic coupling constant below 250 K, and a positive magnetoelastic coupling constant at higher temperatures, as shown in Fig. 5. These two contributions are compensated about 250 K, where the anisotropic magnetostriction becomes zero. Similar magnetostriction compensation has been reported for $Nd_6Fe_{13}Si$ about its spin-reorientation temperature, $T_{SR} = 115\,K$ [11]. Concerning opposite sign of the crystal field anisotropy of $Pr^{3\pm}$ ions at 16l and 8f sites [6], one may attribute negative contribution to the Pr(16l) sublattice that should be dominant at lower temperatures.

Fig. 5 shows that as the temperature approaches to $T_c = 320\,K$, magnetostriction tends to zero due to the natural decrease of the magnetization by thermal fluctuations.

4. Summary

Magnetostriction and linear thermal expansion of $Pr_6Fe_{11}Ga_3$ alloy were reported. The results show that:

(i) Short-range ferromagnetic and long-range ferrimagnetic phases are accompanied by the pronounced spontaneous magnetostrictions, where a sequence of two increasing steps is appeared in the λ_{ex}, with decreasing temperatures.
(ii) Magnetostriction is small at low fields, which is due to the domain wall pinning centers at the Pr crystallographic sites.
(iii) Isofield curves of the magnetostriction pass through a minimum below 250 K, a maximum about 280 K, then approach to zero with increasing temperatures. The magnetostriction compensation about 250 K may arise from the difference in the magnetoelastic coupling constants of $Pr_6Fe_{11}Ga_3$.

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