Magnetostriction and anisotropy compensation in $Tb_xDy_{1-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys

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Structure and magnetostriction of $Dy_{0.7-x}Tb_xPr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ ($0 \leqslant x \leqslant 0.70$) alloys have been studied. The easy magnetization direction of the Laves phase in the $Dy_{0.7-x}Tb_xPr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys with $0 \leqslant x < 0.20$ lies along $\langle 100 \rangle$ axis, while that of the Laves phase in those alloys with $0.25 \leqslant x \leqslant 0.70$ lies along $\langle 111 \rangle$ axis, as determined by means of x-ray crystallography study. When x is increased from 0.15 to 0.25, the change of the easy magnetization direction from $\langle 100 \rangle$ to $\langle 111 \rangle$ axis is detected also by Mössbauer spectra, in good agreement with the results of x-ray crystallography. The Laves phase $Tb_{0.25}Dy_{0.45}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ has a large spontaneous magnetostriction ($\lambda_{111} \approx 1850$ ppm) and a low anisotropy at room temperature, which could make it a good candidate material for magnetostriction applications. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1644057]

For the purpose of application of magnetostrictive materials, besides the factor of their cost, a low magnetocrystalline anisotropy and a large magnetostriction are requested. $Tb_{0.27}Dy_{0.73}Fe_2$ (Terfenol-D) which exhibits a relatively low magnetocrystalline anisotropy while maintaining a large magnetostriction at room temperature has been intensively investigated.[1–4] It has been widely applied in acoustic transducers, sensors, actuators, etc. However, the main raw materials of Terfenol-D are expensive Tb and Dy. It would be of considerable benefit to applications, if a magnetostrictive compound was found, based on the light rare-earths, like Pr, which was much cheaper than Tb and Dy. The single-ion model indicated that the giant magnetostriction of RFe_2 (R = rare earth) is due mainly to the rare-earth ion.[5,6] It was predicted that a Pr ion generates a larger magnetostriction than a Tb or Dy ion. A large amount of work has been done to substitute Pr for Tb or Dy in Terfenol-D.[7–9] However, in most of the previous work, the Pr substitution has been limited to about 10 at. % of the rare-earth component, since beyond 20 at. % substitution, an unanticipated noncubic structure appears. A small amount of boron was introduced to inhibit the appearance of the noncubic phase.[10,11] In this letter, a composition anisotropy compensating point is found in the $Dy_{0.7-x}Tb_xPr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy system, as confirmed by means of the Mössbauer effect and x-ray crystallography study. The linear anisotropy magnetostriction $\lambda_a = \lambda_\parallel - \lambda_\perp$ of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys is studied as well.

All polycrystalline samples of $Dy_{0.7-x}Tb_xPr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys with $x = 0$, 0.10, 0.15, 0.20, 0.25, 0.40, 0.55, and 0.70 were prepared by arc melting of the appropriate constituent metals in a high-purity argon atmosphere. The purities of the constituents are 99.9% for Tb, Dy, Pr, and B, and 99.8% for Fe. The ingots were homogenized at 700 °C for 7 days in a high-purity argon atmosphere. Temperature dependencies of ac initial susceptibility χ_{ac} were recorded at $H = 160$ A/m. Mössbauer analyses were made by a MS-500 spectrometer with the transmission geometry at RT. The source was ^{57}Co in a Pd matrix. The spectra were calibrated with an α-Fe sample and fitted using a standard program. X-ray diffraction (XRD) data were recorded at RT with Cu K_α radiation in a D/max-2500pc diffractometer. In order to investigate the peak splitting induced by the spontaneous magnetostriction, a high-precision step scanning was performed for (440) peaks of XRD. The magnetostriction was measured RT either parallel or perpendicular to the applied field using a standard strain gauge technique.[10,11]

XRD patterns of homogenized $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys are shown in Fig. 1. All the homogenized alloys consist predominantly of the cubic Laves phase with a $MgCu_2$-type structure, with minor impurity phases (i.e., rare-earth phases) and $(Tb,Dy,Pr)_2(Fe,B)_{17}$ phase with a rhombohedral Th_2Zn_{17}-type structure. In the

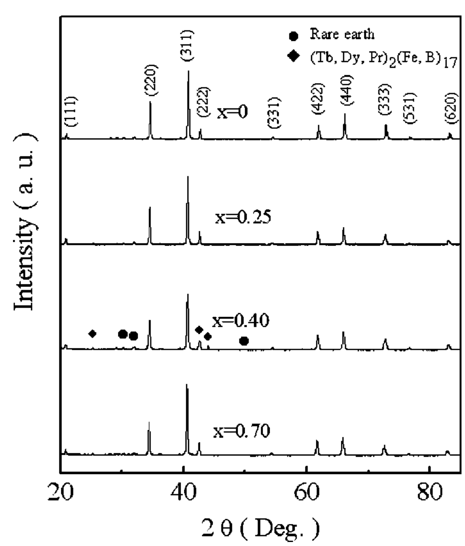

FIG. 1. XRD patterns of homogenized $Dy_{0.7-x}Tb_xPr_{0.3}$ $(Fe_{0.9}B_{0.1})_{1.93}$ alloys. (h,k,l) of the Laves phase was indexed.

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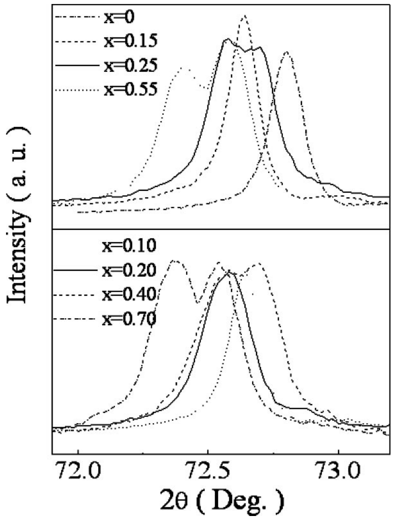

FIG. 2. Profiles of the (440) line of the $Dy_{0.7-x}Tb_xPr_{0.3}$ $(Fe_{0.9}B_{0.1})_{1.93}$ cubic Laves phase.

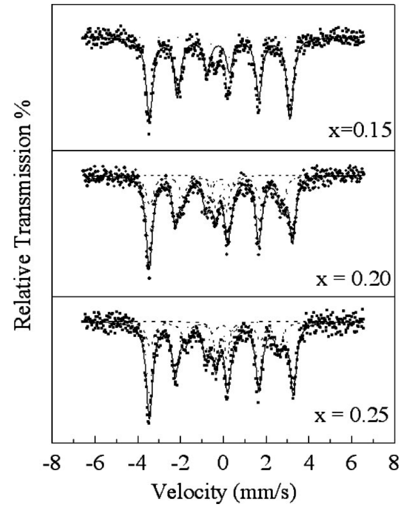

FIG. 3. Mössbauer spectra at RT for $Dy_{0.7-x}Tb_xPr_{0.3}$ $(Fe_{0.9}B_{0.1})_{1.93}$ alloys.

temperature dependence of the ac initial susceptibility χ_{ac} of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys, an anomaly is observed, at about 9 K, which corresponds to the Curie temperature T_C of the minor phase β-Pr(Tb, Dy) with a fcc structure.[12] The amount of $(Tb,Dy,Pr)_2(Fe,B)_{17}$ phase in the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys (except for $x=0.40$) is very small, but changes slightly with x. Only for $x=0.40$, its appearance becomes evident, so that its Curie temperature ($T_C=272$ K) in $Tb_{0.4}Dy_{0.3}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys has been detected in ac initial susceptibility measurements (χ_{ac}–T curves are not shown here).

It is well known that the spontaneous magnetostriction leads to distortion of the crystal structure when a magnetic material is cooled down below its Curie temperature. On the other hand, a rhombohedral or tetrahedral distortion led by magnetostriction can be identified, and the magnetostriction coefficient can be obtained by x-ray crystallography study. In the present work, x-ray step scanning was performed for the (440) XRD line of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys to detect the crystal structure distortion. The profiles of the (440) line after eliminating the effect of the $K_{\alpha 2}$ radiation are represented in Fig. 2. When $0\leqslant x\leqslant 0.20$, each of the (440) lines is a single peak, suggesting that the easy magnetization direction (EMD) of the Laves phase of those alloys lies along $\langle 100\rangle$. Evidently, the tetrahedral distortion, which originated from magnetostriction ($\lambda_{100}\approx 0$), is too small to be observed by XRD. When $0.25\leqslant x\leqslant 0.70$, the double splitting of the (440) lines is clearly observed, indicating that the EMD of the Laves phase in those alloys lie along $\langle 111\rangle$. This is because the magnetostriction leads to a large rhombohedral distortion in those $(Tb,Dy,Pr)(Fe,B)_2$ compounds. The composition anisotropy compensation point is in the range of $0.20\leqslant x\leqslant 0.25$, within which the large rhombohedral distortion disappears.

^{57}Co/Pd Mössbauer spectra for the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys near the anisotropy compensating point were recorded at RT, as shown in Fig. 3. The scattered points are experimental data, the solid lines are gross spectra, and the other lines are subspectra. It can be seen that every Mössbauer spectrum of the alloys consists of one or two six-line subspectra and a two-line subspectrum. The six-line subspectra correspond to the Laves phases, while the two-line ones correspond to a paramagnetic phase that should be attributed to the $(Tb,Dy,Pr)_2(Fe,B)_{17}$ phase in the present alloys. The Curie temperature ($T_C=272$ K) of this phase is little lower than room temperature. The $Tb_{0.15}Dy_{0.55}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy shows only one simple six-line subspectrum, indicating the EMD of the Laves phase lies along $\langle 100\rangle$ axis corresponding to the tetrahedral distortion. The spectrum of the $Tb_{0.25}Dy_{0.45}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy can be decomposed into two six-line subspectra with intensity ratio 3:1, which are characteristic of an EMD along $\langle 111\rangle$ direction, corresponding to rhombohedral distortions. The EMD of the $Tb_{0.2}Dy_{0.5}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy lies along the $\langle 100\rangle$ axis as determined by x-ray step scanning, but its Mössbauer spectrum must be fitted to two six-line subspectra instead of a simple six-line subspectrum. This may be due to an instability in this alloy, with the spins ready to reorient into the $\langle 111\rangle$ direction, just as in Terfenol-D. Atzmony *et al.*[13] also found this kind of intermediate types of spectra, which can be well understood by a two-sublattice mean-field theory.[14,15]

For the Laves phase with EMD lying along the $\langle 111\rangle$ axis, the spontaneous magnetostriction λ_{111} can be obtained from the doubly split (440) XRD line in Fig. 2, using the following equation:[10]

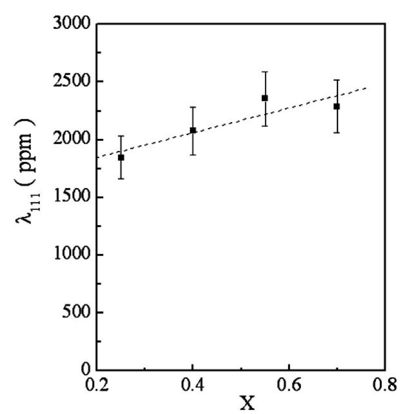

FIG. 4. Concentration dependence of the spontaneous magnetostriction λ_{111} of the $Dy_{0.7-x}Tb_xPr_{0.3}$ $(Fe_{0.9}B_{0.1})_{1.93}$ cubic phase.

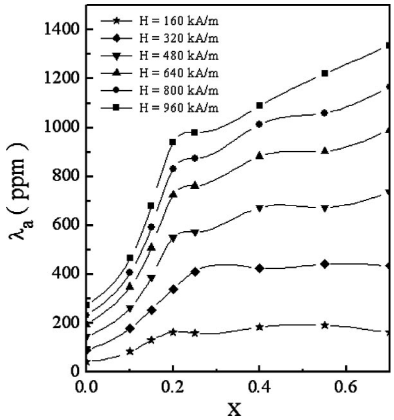

FIG. 5. Linear anisotropy magnetostriction λ_a at different applied fields for polycrystalline $Dy_{0.7-x}Tb_xPr_{0.3}$ $(Fe_{0.9}B_{0.1})_{1.93}$ alloys at RT.

$$\lambda_{111}=2\frac{d_{440}-d_{4\bar{4}0}}{d_{440}+d_{4\bar{4}0}}, \tag{1}$$

where d_{440} and $d_{4\bar{4}0}$ denote the crystallographic plane distances in pseudocubic indices (*hkl*). The composition dependence of the spontaneous magnetostriction λ_{111} is shown in Fig. 4. It can be seen that λ_{111} increases with increasing Tb content in the alloys, in accordance with the observation that $TbFe_2$ has a larger λ_{111} than $DyFe_2$.[5,6,16] Compared with Terfenol-D ($\lambda_{111}\approx 1600$ ppm), the $Tb_{0.25}Dy_{0.45}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy contains 30 at. % Pr (that is much cheaper than Dy), but has a larger magnetostriction ($\lambda_{111}\approx 1850$ ppm). Furthermore, this alloy, which is near the anisotropy compensation point, should have a small anisotropy similar to Terfenol-D. Therefore, it may be a good, practical magnetostriction material. When $x\leqslant 0.20$, the EMD of the Laves phase lies along the $\langle 100\rangle$ axis instead of the $\langle 111\rangle$ axis and, thus the magnetostriction constant λ_{111} can not be obtained anymore using our x-ray method. The linear anisotropic magnetostriction $\lambda_a=\lambda_\parallel-\lambda_\perp$ at RT of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ polycrystalline alloys is shown in Fig. 5. Its behavior is similar to the magnetostriction of $(Tb_{1-x}Dy_x)Fe_2$ alloys.[16]

In conclusion, the composition anisotropy compensation has been realized near $x=0.20$ for the $Dy_{0.7-x}Tb_xPr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys. The Laves phase $Tb_{0.25}Dy_{0.45}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ has a large spontaneous magnetostriction ($\lambda_{111}\approx 1850$ ppm) and a low anisotropy at RT which could make it a good candidate material for magnetostriction application.

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