Spin Orientation and Spontaneous Magnetostriction of Multicomponent $Tb_xDy_{1-x-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves Phases

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***Abstract*—The spin orientation and spontaneous magnetostriction of multicomponent $Tb_xDy_{1-x-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ ($0.10 \leq x \leq 0.25, 0.30 \leq y \leq 0.60$) Laves phases were studied by step-scanning their (440) X-ray diffraction lines. The easy magnetization direction (EMD) of the Laves phases changed from the $\langle 100 \rangle$ to $\langle 111 \rangle$ axis when x was increased from 0.10 to 0.25. With increasing y, the EMD of $Tb_{0.15}Dy_{0.85-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ and $Tb_{0.2}Dy_{0.8-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases reoriented from the $\langle 100 \rangle$ to $\langle 111 \rangle$ axis. The magnetostriction coefficient λ_{111} of $Tb_xDy_{1-x-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases increased with increasing x (or y) when y (or x) was kept constant.**

***Index Terms*—Laves phases, magnetostriction, spin orientation, X-ray diffraction.**

I. Introduction

PSEUDOBINARY $MgCu_2$-type $RR'Fe_2$ alloys ($R, R' \equiv$ rare earths) have good prospects as practical magnetostrictive materials due to their large spontaneous linear magnetostriction and low magnetocrystalline anisotropy at room temperature. Terfenol-D ($Tb_{0.30}Dy_{0.70}Fe_{1.92}$), which exhibits giant magnetostriction of ~1200 ppm at a low magnetic field of ~80 kA/m, is the most widely used magnetostrictive material to date [1]. Nevertheless, Terfenol-D suffers from high cost because of alloying high contents of heavy rare-earths Tb and Dy with Fe. Wu *et al.* studied a multicomponent pseudobinary $Tb_xDy_{1-x-y}Pr_yFe_2$ system by using a cost-effective light rare-earth Pr in substitution for Tb and Dy [2]. However, the Pr content was limited to a low value of 0.20, beyond which unanticipated noncubic structure occurred.

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We have recently shown that the substitution of B to such a system is beneficial to the formation of Laves phases with increased Pr content [3], [4]. In this paper, the spin orientation and spontaneous magnetostriction of multicomponent $Tb_xDy_{1-x-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ ($0.10 \leq x \leq 0.25$, $0.30 \leq y \leq 0.60$) Laves phases are investigated to reveal their composition anisotropy compensation effect.

II. Experiment

Polycrystalline samples of $Tb_xDy_{1-x-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ with $x = 0.10$, 0.15, 0.20, and 0.25 and $y = 0.30$, 0.40, 0.50, and 0.60 were prepared by arc-melting the appropriate constituent metals in a high purity argon atmosphere. The purity of Tb, Dy, Pr, and B was 99.9%, whlist that of Fe was 99.8%. The ingots were homogenized at 700 °C for 7 days in an argon atmosphere. X-ray diffraction (XRD) was implemented at room temperature with CuK_α radiation in a Riguku D/max-2500 pc diffractrometer equipped with a graphite monochromator. In order to study the easy magnetization direction (EMD) and magnetostriction coefficient λ_{111} of the Laves phases, a high-precision XRD step scanning was performed on powdered samples from 72° to 73° for the (440) peaks. The effect of the $K_{\alpha 2}$ radiation was eliminated. The XRD peaks were fitted using the fitting function PearsonVII provided by Jade 6.5 XRD analytical software (Materials Data, Inc., Livermore, CA).

III. Results and Discussions

The step-scanned XRD profiles for $Tb_{0.10}Dy_{0.90-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases with $y = 0.30$, 0.40, 0.50, and 0.60 are shown in Fig. 1. All profiles exhibit a sharp single peak with 2θ of ~72.6° and with no observable split-up in peak. This suggests that the EMD of these Laves phases lies along the $\langle 100 \rangle$ axis [5], and the distortion in crystal structure due to magnetostriction is minimal (i.e., $\lambda_{100} \sim 0$). The step-scanned profiles for $Tb_{0.15}Dy_{0.85-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases are plotted in Fig. 2. The (440) XRD lines of the Laves phases remain essentially a sharp single peak for $y = 0.30$ and 0.40 but are doubly split and partially overlap to form a broadened peak when y is increased to 0.50 and 0.60. The split peaks located at 72.52° and 72.62° are for $y = 0.50$, while those seen at 72.41° and 72.68° are for $y = 0.60$. This is because the EMD of these Laves phases is along the $\langle 111 \rangle$ axis, and a large rhombohedral distortion in the crystal structure occurs. That is, the magnetostriction distorts the cubic system into a rhombohedral system, and the (440) lines

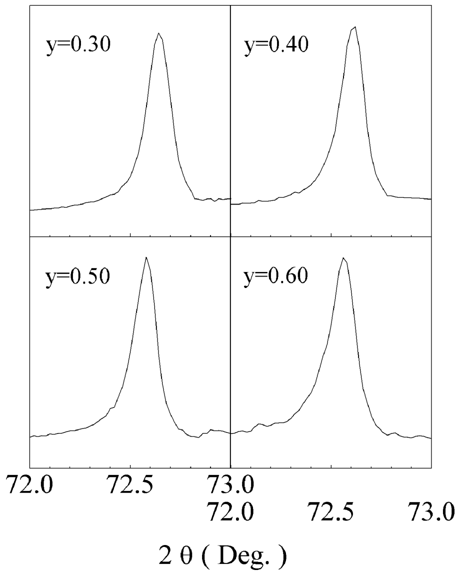

Fig. 1. Step-scanned XRD profiles for $\mathrm{Tb}_{0.10}\mathrm{Dy}_{0.90-y}\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ Laves phases.

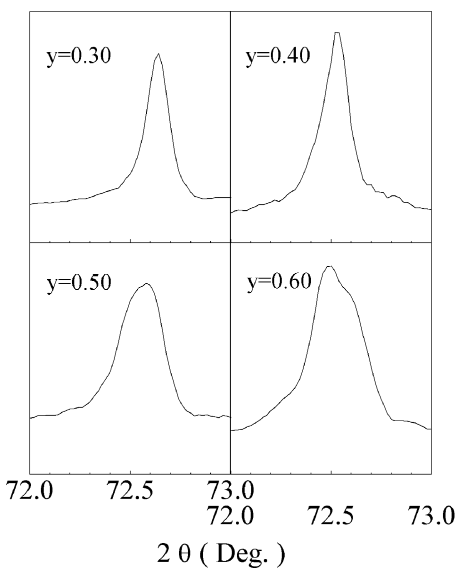

Fig. 2. Step-scanned XRD profiles for $\mathrm{Tb}_{0.15}\mathrm{Dy}_{0.85-y}\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ Laves phases.

of the cubic Laves phases are split into the (242) and $(20\ \overline{2})$ lines of the rhombohedral system corresponded to the (440) and $(4\ \overline{4}\ 0)$ lines of a pseudo-cubic system. Fig. 3 shows the step-scanned profiles for $\mathrm{Tb}_{0.25}\mathrm{Dy}_{0.75-y}\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ Laves phases. The double-split (440) lines of the Laves phases are clearly observed in all spectra, reflecting the presence of a large magnetostriction along the $\langle 111\rangle$ EMD of these Laves phases. The split peaks and y values are identified as follows: peaks at 72.58° and 72.71° are for $y = 0.30$; peaks at 72.46° and 72.60° are for $y = 0.40$; peaks at 72.40° and 72.55° are for $y = 0.50$; and peaks at 72.34° and 72.51° are for $y = 0.60$. No step-scanned profiles are presented for $\mathrm{Tb}_{0.20}\mathrm{Dy}_{0.80-y}\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ Laves phases as they have been reported in a previous article [4]. Nevertheless, a similar change in the EMD from the $\langle 100\rangle$ to $\langle 111\rangle$ axis has been found.

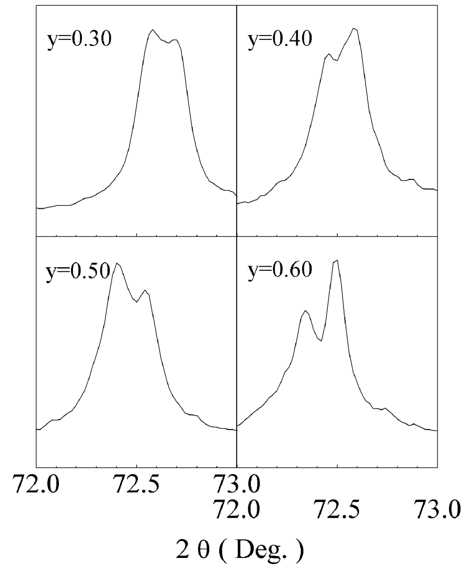

Fig. 3. Step-scanned XRD profiles for $\mathrm{Tb}_{0.25}\mathrm{Dy}_{0.75-y}\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ Laves phases.

TABLE I
SUMMARY OF THE EASY MAGNETIZATION DIRECTION (EMD) AND MAGNETOSTRICTION COEFFICIENT λ_{111} OF MULTI-COMPONENT $\mathrm{Dy}_{1-x-y}\mathrm{Tb}_x\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ $(0.10 \le x \le 0.25, 0.30 \le y \le 0.60)$ LAVES PHASES

EMD / λ_{111} (ppm)	x=0.10	x=0.15	x=0.20	x=0.25
y=0.3	<100> / –	<100> / –	<100> / –	<111> / 1538
y=0.4	<100> / –	<100> / –	<111> / 1182	<111> / 1613
y=0.5	<100> / –	<111> / 1152	<111> / 1459	<111> / 1842
y=0.6	<100> / –	<111> / 1536	<111> / 1689	<111> / 1994

Based on the observed double-split (440) lines (Figs. 1–3 and [4, Fig. 3]), the magnetostriction coefficient λ_{111} of multicomponent $\mathrm{Tb}_x\mathrm{Dy}_{1-x-y}\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ $(0.10 \le x \le 0.25, 0.30 \le y \le 0.60)$ Laves phases was calculated using the following equation [3]:

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

where d_{440} and $d_{4\bar{4}0}$ denote the crystallographic plane distances in pseudo-cubic indexes (*hkl*). The results are summarized in Table I, together with those of the EMD. For a given y, it is clear that the EMD of $\mathrm{Tb}_x\mathrm{Dy}_{1-x-y}\mathrm{Pr}_y(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_{1.93}$ Laves phases changes from the $\langle 100\rangle$ to $\langle 111\rangle$ axis when x is increased from 0.10 to 0.25 due to the anisotropy compensation of Tb^{3+} for Dy^{3+}. For $x = 0.15$ and 0.20, the EMD of the Laves phases reorients from the $\langle 100\rangle$ to $\langle 111\rangle$ axis when y is increased from 0.30 to 0.60 because of the anisotropy compensation between Dy^{3+} and Pr^{3+}. This can be realized as $\mathrm{Dy}_{1-x}\mathrm{Pr}_x\mathrm{Fe}_2$ is a composition anisotropy compensation system, as phenomenologically proved by a single-ion approach [4]. The magnetostric-

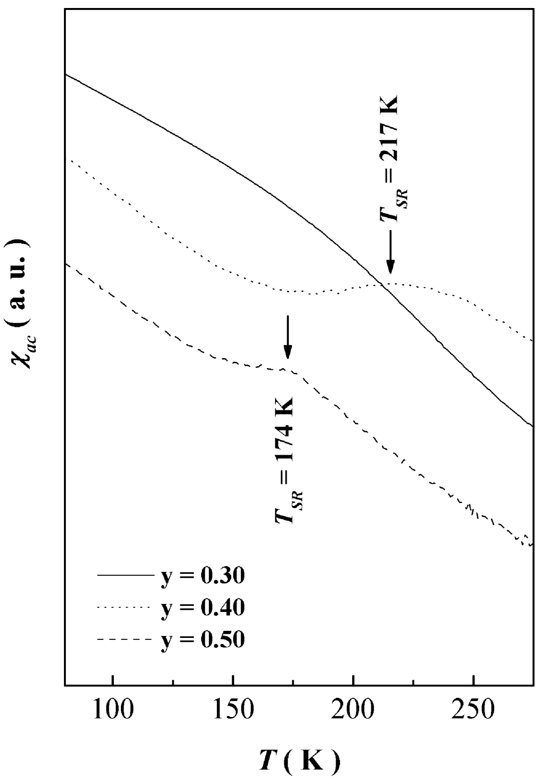

Fig. 4. Temperature dependence of ac initial susceptibility for $Tb_{0.2}Dy_{0.8-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases.

tion coefficient λ_{111} of $Tb_xDy_{1-x-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases increases with increasing x (or y) when y (or x) is an invariant. This is because λ_{111} of $TbFe_2$ (or $PrFe_2$) is larger than that of $DyFe_2$.

The temperature dependence of ac initial susceptibility (χ_{ac}) for $Tb_{0.2}Dy_{0.8-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases with $y = 0.30$, 0.40, and 0.50 was measured with a magnetic field of 160 A/m, a frequency of 300 Hz, and a range of temperatures from 80 K to 273 K in order to determine their spin reorientation temperatures (T_{SR}) for better understanding of the spin orientation effect. The detected anomalies at 174 K and 217 K for $Tb_{0.2}Dy_{0.3}Pr_{0.5}(Fe_{0.9}B_{0.1})_{1.93}$ and $Tb_{0.2}Dy_{0.4}Pr_{0.4}(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases, respectively, are their T_{SR} [6]. The observation agrees with our reported XRD results ([4, Fig. 3] and Table I) in that the EMD of the Laves phases is along the $\langle 111 \rangle$ axis at room temperature. T_{SR} is not observed on $Tb_{0.2}Dy_{0.5}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ Laves phase in the temperature range of measurement; it should be higher than room temperature as the EMD of this Laves phase lies along the $\langle 100 \rangle$ axis at room temperature (Table I). It is interesting to note that T_{SR} of the $Tb_{0.2}Dy_{0.8-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phase for $0.30 < y < 0.40$ is close to room temperature, similar to the case of Terfenol-D. Thus, they should also have low magnetocrystalline anisotropy at room temperature.

IV. Conclusion

The spin orientation and spontaneous magnetostriction of multicomponent $Tb_xDy_{1-x-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases have been investigated by XRD. Results show that the anisotropy of Tb^{3+} and/or Pr^{3+} can compensate for that of Dy^{3+}. The magnetostriction coefficient λ_{111} of the Laves phases increases with increasing x (or y) when y (or x) remains unchanged. The single $Tb_{0.2}Dy_{0.8-y}Pr_y(Fe_{0.9}B_{0.1})_{1.93}$ Laves phase with $0.30 < y < 0.40$ has large magnetostriction and low magnetocrystalline anisotropy at room temperature. It may be a good practical magnetostrictive material, which is less expensive in comparison with Terfenol-D.

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