

Magnetostriction of $Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.9}B_{0.1})_2$ ($x = 0 - 0.4$) compounds

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Magnetostriction of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ ($x=0-0.4$) compounds

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The structural and magnetic properties of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ ($x=0, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35,$ and 0.4) compounds are investigated. The compounds with $0 \leq x \leq 0.25$ are found to stabilize in single cubic Laves phase structures. Small quantities of the $(\text{Tb}, \text{Ho}, \text{Pr})_2\text{Fe}_{14}\text{B}$ phase and rare-earth phases for $x \geq 0.3$ and also the $(\text{Tb}, \text{Ho}, \text{Pr})(\text{Fe}, \text{B})_3$ phase for $x \geq 0.35$ are observed along with the Laves phase. The saturation magnetization and Curie temperature are found to decrease with Pr content. However, the magnetostriction (at 10 kOe) is found to increase with Pr content. The $x=0.15$ compound is found to exhibit a possible anisotropy compensation between the (Tb/Ho) and Pr sublattices. The easy magnetization direction (EMD) is found to rotate toward $\langle 113 \rangle$ as the Pr content increases, up to $x=0.25$, and for $x \geq 0.35$ there is a tendency for the EMD to shift toward $\langle 111 \rangle$. © 2008 American Institute of Physics. [DOI: 10.1063/1.2828588]

I. INTRODUCTION

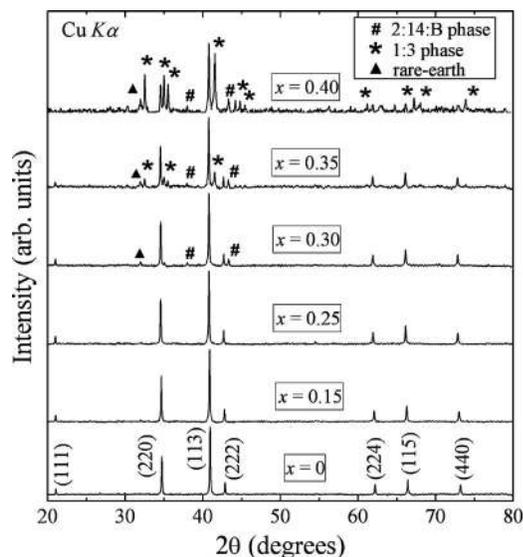
The binary C15 cubic Laves phase $R\text{Fe}_2$ (R = rare-earth elements) compounds are known to exhibit giant magnetostriction values and large magnetocrystalline anisotropy at room temperature.^{1,2} Clark¹ calculated spontaneous magnetostriction (λ_{111}) values for $R\text{Fe}_2$ compounds at 0 K, and the values of PrFe_2 , TbFe_2 , DyFe_2 , SmFe_2 , NdFe_2 , and HoFe_2 were 5600×10^{-6} , 4400×10^{-6} , 4200×10^{-6} , -3200×10^{-6} , 2000×10^{-6} , and 1600×10^{-6} , respectively. Savage *et al.*³ reported that the saturation magnetostriction (λ_s) of the anisotropy compensated compound $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ is 1000×10^{-6} . Williams and Koon⁴ reported that $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$, another anisotropy compensated pseudobinary compound, has a saturation magnetostriction of 325×10^{-6} at room temperature. PrFe_2 and NdFe_2 compounds cannot be synthesized through conventional preparation methods¹ though PrFe_2 has been synthesized by high pressure methods.⁵ Recently, Hari Babu *et al.*⁶ reported a magnetostriction ($\lambda_{\parallel} - \lambda_{\perp}$) of 2220×10^{-6} in a $\text{Sm}_{0.88}\text{Nd}_{0.12}\text{Fe}_{1.93}$ compound. Ren *et al.*⁷ reported the anisotropy compensation between Dy^{3+} and Pr^{3+} ions in $\text{Tb}_{0.2}\text{Dy}_{0.8-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_{1.93}$ compounds. The easy magnetization direction (EMD) in PrFe_2 has been reported to be along the $\langle 111 \rangle$ direction at room temperature.⁵ It has also been reported that the EMD of the TbFe_2 and HoFe_2 are along $\langle 111 \rangle$ and $\langle 100 \rangle$, respectively, accompanied by rhombohedral and tetrahedral distortions, respectively, at room temperature.¹ In the present work, we report the anisotropy compensation and magnetostriction in

$\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compounds. Boron was substituted at the Fe site to stabilize the compounds in the Laves phase.⁷⁻⁹

II. EXPERIMENTAL DETAILS

All the $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ ($x=0, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35,$ and 0.4) compounds were prepared by arc melting the constituent elements (R : 99.9% pure; Fe and B: 99.95% pure) in an arc furnace under argon atmosphere. The ingots were sealed in quartz ampoules under $\sim 10^{-6}$ mbar pressure and were annealed at 900 °C for 7 days. Powder x-ray diffraction (XRD) patterns of the compounds were taken using a PANalytical (X'pert PRO) x-ray diffractometer employing Cu $K\alpha$ radiation. The room temperature magnetization measurements up to a magnetic field of 12 kOe were carried out using a PAR-155 vibrating sample magnetometer. The Curie temperature (T_C) values were estimated using the observation of phase transitions in the specific heat measurements. The room temperature magnetostriction measurements up to a magnetic field of 10 kOe were carried out on the rod shaped (4 mm diameter and 15 mm long) samples of the compounds, employing the strain gauge method.¹⁰ In order to determine the EMD of the above compounds, samples were prepared using the fine powders of the compounds mixed with an epoxy resin. These powders were aligned in the presence of a magnetic field of 20 kOe, and the epoxy resin was allowed to cure for 30 min.

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FIG. 1. XRD patterns of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compounds.

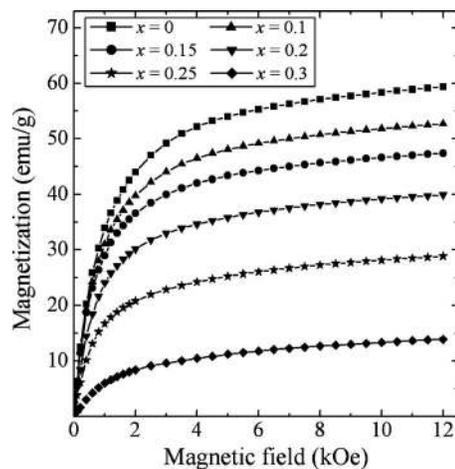
III. RESULTS AND DISCUSSION

The powder XRD patterns of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ ($x=0, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35$, and 0.4) compounds are shown in Fig. 1. The compounds with $0 \leq x \leq 0.25$ are found to stabilize in the C15 cubic Laves phase crystal structure. Small quantities of $(\text{Tb}, \text{Ho}, \text{Pr})_2\text{Fe}_{14}\text{B}$ (2:14:B phase) and rare-earth phases appeared for $x \geq 0.3$ compounds in addition to the cubic Laves phase. In addition to all the above phases, the $(\text{Tb}, \text{Ho}, \text{Pr})(\text{Fe}, \text{B})_3$ phase (1:3 phase) with a PuNi_3 -type crystal structure is also seen to grow for $x \geq 0.35$ compounds. The XRD pattern of the $x=0.4$ compound revealed the presence of a large quantity of 1:3 phase amongst the other phases. Therefore, the boron substitution seems to prevent the formation of the additional phases only up to $x=0.25$. The lattice parameter a is found to significantly increase with the Pr content up to $x=0.25$ (Table I), in accordance with the larger size of the Pr^{3+} ion than the Ho^{3+} ion.

The magnetization curves of the $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ ($x=0, 0.1, 0.15, 0.2, 0.25$, and 0.3) compounds at room temperature are shown in Fig. 2. The saturation magnetization (M_s) values were calculated using the Honda plots (M versus $1/H$) and are listed in Table I. M_s is found to decrease with increasing Pr content. The total magnetic moment in the $(\text{Tb}, \text{Ho})\text{Fe}_2$ compound is dominated by the Ho and Tb (heavy rare earths) moments that are antiparallel to the Fe moment. On the other hand, Pr

TABLE I. Lattice parameter (a), saturation magnetization (M_s), and Curie temperature (T_C) values of $\text{Tb}_x\text{Ho}_{0.75-x}\text{Pr}_{0.25}(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compounds.

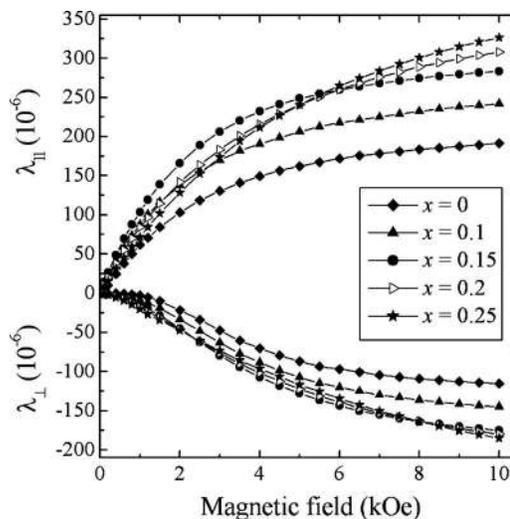
Sample No.	x	a (± 0.002 Å)	M_s (emu/g)	T_C (K)
1	0	7.305	64.83	612
2	0.1	7.316	57.52	607
3	0.15	7.324	51.67	604
4	0.2	7.332	43.60	
5	0.25	7.338	33.01	599
6	0.3	7.340	16.27	

FIG. 2. Magnetization curves of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compounds at room temperature.

is a light rare earth, and the magnetic moment of Pr is antiparallel to that of Tb/Ho moments,¹¹ causing a decrease in the net moment, at least for the low concentrations of Pr as, the compounds with large Pr content could not be prepared in a single cubic Laves phase structure.

The Curie temperature (T_C) is seen to decrease with increasing Pr content up to $x=0.25$ (Table I), in accordance with smaller de Gennes factor of the Pr^{3+} ion ($0.8\mu_B$) compared to that of the Ho^{3+} ion ($4.5\mu_B$).¹²

The room temperature magnetostriction curves of the $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ ($x=0, 0.1, 0.15, 0.2$, and 0.25) compounds are shown in Fig. 3. The parallel magnetostriction (λ_{\parallel}) and perpendicular magnetostriction (λ_{\perp}) correspond to the strain when the magnetic field was applied along and perpendicular to the axis of the rod, respectively. The λ_{\parallel} and λ_{\perp} values (at 10 kOe) are found to increase significantly with increasing Pr content, in accordance with the larger predicted λ_{111} value of the PrFe_2 (5600×10^{-6}) compound to that of the HoFe_2 (1600×10^{-6}) compound at 0 K.¹ Relatively large λ_{\parallel} and λ_{\perp} are observed at low magnetic fields for the $x=0.15$ compound compared to the other compounds in-

FIG. 3. Magnetostriction curves of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compounds at room temperature.

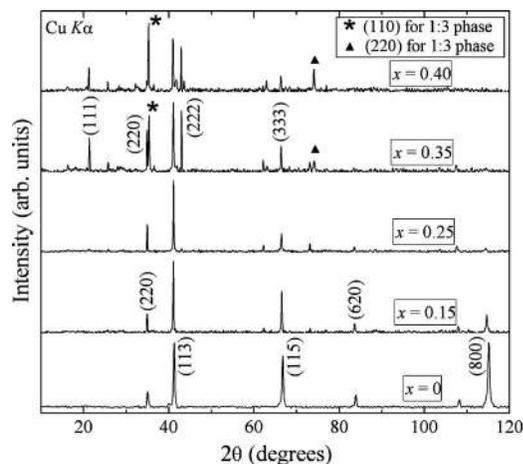


FIG. 4. XRD patterns of magnetically aligned powders of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compounds.

investigated here. Clark¹ reported that the signs of first order anisotropy constant (K_1) of TbFe_2 , HoFe_2 , and PrFe_2 are negative, positive, and positive, respectively. He also reported that the signs of second order anisotropy constant (K_2) of TbFe_2 , HoFe_2 , and PrFe_2 are positive, positive, and negative, respectively. Thus, the compensation between the anisotropies of the (Tb/Ho) and Pr sublattices can be expected at room temperature. Moreover, the $\text{Tb}_{0.1}\text{Ho}_{0.9}\text{Fe}_2$ compound is known as a nearly anisotropy compensated compound. Therefore, the compensation at $x=0.15$ is mainly due to the K_2 values of Ho and Pr sublattices. Ren *et al.*^{7,13} reported anisotropy compensation between the Dy and Pr sublattices in $\text{Tb}_{0.2}\text{Dy}_{0.8-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_{1.93}$ and $\text{Dy}_{1-x}\text{Pr}_x(\text{Fe}_{0.35}\text{Co}_{0.55}\text{B}_{0.1})_2$ compounds. The compensation in the above compounds is in accordance with the opposite signs of the K_2 values of Dy and Pr sublattices, though they both have the positive K_1 values. In a similar way, the present $\text{Tb}_{0.1}\text{Ho}_{0.75}\text{Pr}_{0.15}(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compound is known to have the compensation of the K_2 values of Ho and Pr sublattices. The λ_s value of the above compound (305×10^{-6}) is comparable to that of $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$ (325×10^{-6}).⁴ In spite of the suppression of the magnetostriction by B, the presence of Pr is seen to contribute to an effective increase of magnetostriction.^{14,15}

In order to determine the EMD in the $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ compounds, XRD patterns were taken on the magnetically aligned powders of the above compounds employing $\text{Cu } K\alpha$ radiation, and the patterns are shown in Fig. 4. The $\text{Tb}_{0.1}\text{Ho}_{0.9}\text{Fe}_{(0.9}\text{B}_{0.1})_2$ compound is found to have intense reflections corresponding to (113), (115), and (800) planes, indicating lack of axial anisotropy. The EMD of the $\text{Tb}_{0.1}\text{Ho}_{0.9}\text{Fe}_2$ compound at room temperature is in between the $\langle 111 \rangle$ and $\langle 100 \rangle$ directions.¹⁶ On the other hand, the compounds $x=0.15$ and 0.25 are found to have intense reflections corresponding to (113) planes compared with the other reflections, indicating that the EMD is nearly along $\langle 113 \rangle$. For the $x \geq 0.35$ compounds, $\langle 111 \rangle$ reflection was observed along with that corresponding to $\langle 113 \rangle$.

Increasing Pr content (having $\langle 111 \rangle$ as the EMD) could eventually cause the EMD to be along $\langle 111 \rangle$. Thus, the EMD is found to continuously rotate from the $\langle 100 \rangle$ direction ($x=0$) toward the $\langle 111 \rangle$ direction ($x \geq 0.35$) through $\langle 113 \rangle$. On the other hand, the compounds with $x \geq 0.35$ are found to have peaks corresponding to $\langle 110 \rangle$ planes of the 1:3 phase.

IV. CONCLUSIONS

The structural and magnetic properties of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.9}\text{B}_{0.1})_2$ ($x=0, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35$, and 0.4) compounds are presented. The compounds with $0 \leq x \leq 0.25$ are found to stabilize in a single cubic Laves phase structure. The lattice parameter is found to increase with Pr content up to $x=0.25$, in accordance with a larger size of the Pr^{3+} ion than the Ho^{3+} ion. The M_s and T_C are found to decrease with Pr content, in accordance with the reports that the moment of the Pr is antiparallel to that of Tb/Ho and the lower de Gennes factor of Pr^{3+} , respectively. The λ_{\parallel} and λ_{\perp} values (at 10 kOe) are found to increase with Pr content due to the large predicted magnetostriction value of PrFe_2 . The anisotropy compensation is seen in the $x=0.15$ compound due to the K_2 values of Pr and Ho sublattices. The EMD is found to rotate toward $\langle 113 \rangle$ as the Pr content increases, up to $x=0.25$, and for $x \geq 0.35$ there is a tendency for the EMD to shift toward $\langle 111 \rangle$.

ACKNOWLEDGMENTS

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