Spin configuration and magnetostrictive properties of Laves compounds $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}(0.10 \leqslant x \leqslant 0.28)$

J. J. Liu[a)]
Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang, 110016, People's Republic of China; International Centre for Materials Physics, Chinese Academy of Sciences, Shenyang 110016, People's Republic of China; and Department of Applied Physics, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong

W. J. Ren, Z. D. Zhang, D. Li, J. Li, X. G. Zhao, and W. Liu
Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, People's Republic of China and International Centre for Materials Physics, Chinese Academy of Sciences, Shenyang 110016, People's Republic of China

S. W. Or
Department of Applied Physics, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong

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The spin configuration and magnetostrictive properties of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ $(0.10 \leqslant x \leqslant 0.28)$ Laves compounds are investigated. The spin phase diagram for the compounds is constructed to illustrate both the composition dependence of the Curie point T_C and spin reorientation temperature T_{SR} and the arrangement for the easy magnetization direction (EMD). It is found that when x is increased from 0.16 to 0.28, T_C increases from 617 to 623 K, while T_{SR} decreases from 375 to 160 K at which the EMD reorients from $\langle 100 \rangle$ to $\langle 111 \rangle$ axis. The magnetization and magnetostriction of the compounds are measured as a function of applied field. The magnetocrystalline anisotropy constant K_1 and the ratio λ_a/K_1 reach a minimum and a maximum value at $x=0.22$, respectively. The single phase $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ compound has the lowest magnetocrystalline anisotropy, the highest magnetostriction to anisotropy ratio, and a large spontaneous magnetostriction λ_{111} of ~1500 ppm at room temperature. In view of these properties, the compound is a promising magnetostrictive material. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2219344]

I. INTRODUCTION

The cubic Laves compounds $(R,R')Fe_2$ $(R,R' \equiv$ rare earths), particularly the anisotropy-compensated $Tb_{0.27}Dy_{0.73}Fe_2$ (Terfenol-D), have been widely used as actuators and transducers due to their giant magnetostriction and low magnetocrystalline anisotropy at room temperature.[1,2] The low magnetocrystalline anisotropy in Terfenol-D can be ascribed to its spin reorientation temperature T_{SR} near room temperature.[3] The magnetocrystalline anisotropy vanishes at T_{SR}, accompanying a reorientation of the easy magnetization direction (EMD). It would be of considerable benefit to applications if Tb and/or Dy could be totally or partially substituted by low-cost, light rare earths while competitive magnetostrictive properties could still be maintained. In this way, the light rare earth Pr is considered as a good candidate due to the large magnetostriction exhibited by $PrFe_2$ (5600 ppm) as compared with the $TbFe_2$ (4400 ppm) and $DyFe_2$ (4200 ppm) compounds at 0 K, as evidenced by the single-ion model results.[4] However, it remains challenging to synthesize $PrFe_2$ at ambient pressure; in fact, there is a real challenge to preparing Pr-containing compounds with a single Laves phase if the Pr content exceeds 20 at. % of rare earth owing to the large radius of Pr^{3+}.[4–6]

Recently, $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ cubic Laves compounds were synthesized by the addition of small amount of boron as the stabilizing element.[7,8] Besides, there exists a large spontaneous magnetostriction and low magnetocrystalline anisotropy at room temperature in the compounds with $x \sim 0.25$. From the practical viewpoint, it is really necessary to investigate in detail the influence of composition variation on the spin configuration, magnetocrystalline anisotropy, and magnetostrictive properties of the compounds.[9] As a good example, $Tb_{0.27}Dy_{0.73}Fe_2$ and $Tb_{0.3}Dy_{0.7}Fe_2$ show significant differences in magnetocrystalline anisotropy and magnetostrictive properties.[10] It is understood that T_{SR} is crucial for the realization of good magnetostrictive properties. On the other hand, the spin reorientation temperature needs to be near room temperature as close as possible, loading to the small anisotropy, which is important for practical application at low magnetic fields. In addition, if the spin reorientation temperature were too close to the room temperature, the spontaneous magnetostriction and the rhombohedral distortion would disappear rapidly. In this paper, the spin configuration and magnetostrictive properties of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ Laves compounds are investigated in detail, in the x range of 0.10 and 0.28 with a compositional interval $\Delta x=0.03$. The $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy showing optimized properties, which has a suitable spin reorientation temperature $T_{SR}=245$ K, a large magnetostriction coefficient λ_{111}

a)Electronic mail: liujj@imr.ac.cn

≈1500 ppm, and a low magnetocrystalline anisotropy constant K_1 at room temperature, results to be a promising material regarding magnetostriction applications.

II. EXPERIMENTAL PROCEDURE

Polycrystalline samples of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys with x=0.10, 0.13, 0.16, 0.19, 0.22, 0.25, and 0.28 were prepared by using an arc melting with 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 973 K for seven days in a high purity argon atmosphere. X-ray diffraction (XRD) technique was implemented at room temperature with Cu Kα radiation in a Riguku D/max-2500pc diffractrometer equipped with a graphite monochromator. In order to study the EMD and magnetostriction coefficient λ_{111} of the Laves phases, a high-precision XRD step scanning was performed on powdered samples for the (440) peaks. The effect of the $K\alpha_2$ radiation was eliminated. The XRD peaks were fitted using the fitting function PEARSON VII provided by JADE 6.5 XRD analytical software (Materials Data, Inc., Livermore, CA). The ac initial susceptibility temperature dependences were obtained using a superconducting quantum interference device (SQUID) magnetometer to detect the spin reorientation temperature T_{SR}.[11] Magnetization curves, at room temperature, were measured by the SQUID magnetometer at fields up to 50 kOe. The magnetostriction, at room temperature, was measured either in the parallel or perpendicular configuration to the applied field using a standard strain gauge technique.

III. RESULTS AND DISCUSSION

It is well known that the minimum of the magnetocrystalline anisotropy frequently occurs at the spin reorientation temperature T_{SR} with an accompanying magnetization rotation. As for Terfenol-D, the EMD lies along the ⟨111⟩ axis for temperature values greater than T_{SR}, while it lies along the ⟨100⟩ axis for temperature below T_{SR}.[3,12] The ac initial susceptibility temperature dependence $\chi_{ac}(T)$ of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys is shown in the Fig. 1. A significant anomaly is observed in each alloy with $0.16 \leqslant x \leqslant 0.28$, corresponding to a spin reorientation taking place from ⟨100⟩ to ⟨111⟩.[11,12] Such anomaly does not appear in the temperature variation of χ_{ac} for the alloys with $0.10 \leqslant x \leqslant 0.13$ below the maximum available temperature of 380 K, since the spin reorientation of these alloys should take place above 380 K.[9] The phase diagram of the spin configuration for $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases is plotted in Fig. 2, which is obtained from the collection of the spin reorientation temperature T_{SR} (anomaly temperature in Fig. 1), Curie temperature T_C, and easy magnetization direction determined by Mössbauer spectra or XRD method.[7,13–15] The Curie temperature T_C linearly increases as Tb content increases, because the Curie temperature of $TbFe_2$ is larger than that of $DyFe_2$. The spin reorientation temperature T_{SR} decreases from 375 K for the alloy with $x=0.16$ to 160 K for the alloy with $x=0.28$, as Tb content is increased, which can be ascribed to the EMD of $TbFe_2$ that lies along ⟨111⟩ in all

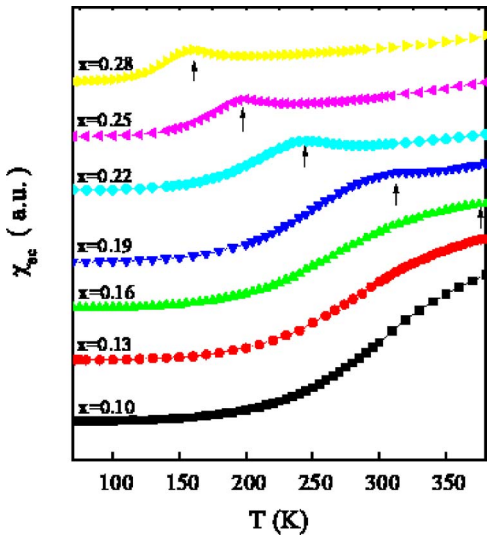

FIG. 1. (Color online) ac initial susceptibility temperature dependence $\chi_{ac}(T)$ of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys. (The arrow indicates spin reorientation temperature T_{SR}.)

the temperature range.[9] Clearly, the Tb-rich alloy is suitable for low temperature applications due to its low T_{SR} and anisotropy at low temperatures, whereas the Tb-poor alloy is preferred for high temperature applications.

The magnetic field dependence of the magnetization at room temperature of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys is shown in Fig. 3. The magnetization for all the alloys studied does not vary notably, owing to the magnetizations of $TbFe_2$ and $DyFe_2$ at room temperature are very close.[16] The magnetocrystalline anisotropy constant K_1 of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ Laves compound is determined by simulating the M-H curves using the approach to the saturation magnetization law as follows:[17]

$$M = M_s\left(1 - \frac{a}{H} - \frac{b}{H^2}\right) + \chi_p H, \tag{1}$$

and the relation[17]

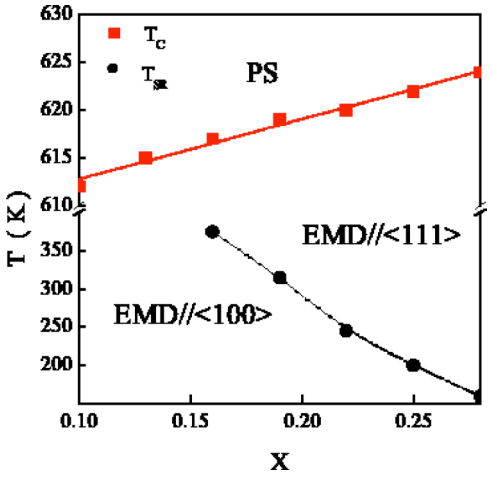

FIG. 2. (Color online) Spin configuration diagram of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ compounds. The squares and circles denote the Curie temperature and the spin reorientation temperature, respectively. PS represents the paramagnetic state.

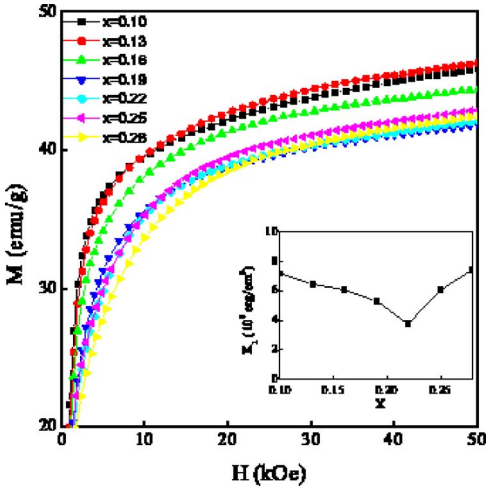

FIG. 3. (Color online) Magnetization curves and magnetocrystalline anisotropy constant K_1, at room temperature of the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys.

$$b = \frac{8}{105}\frac{K_1^2}{\mu_0^2 M_s^2}, \tag{2}$$

where M_s is the saturation magnetization, a and b are constants, χ_p is the susceptibility of the paramagnetic (parallel) magnetization process, and μ_0 is the permeability of the free space. The dependence of the anisotropy constant K_1 on the Tb content x is shown in the inset of the Fig. 3. The anisotropy constant K_1 decreases with increasing Tb content when $0.10 \leqslant x \leqslant 0.22$, then increases with a further increasing x. The $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy has the minimum magnetocrystalline anisotropy at room temperature, in consistent with its $T_{SR}=245$ K, very near to the room temperature.

The magnetic field dependence of the magnetostriction $\lambda_a(=\lambda_\parallel-\lambda_\perp)$ for the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ $(0.10 \leqslant x \leqslant 0.28)$ alloys is shown in Fig. 4. It can been seen that λ_a at the maximum available magnetic field increases with increasing x, which can be due to the increase of λ_{111}.[7] The magnetostriction λ_a for $x=0.22$ is found to be largest at relatively low fields, owing to its lowest anisotropy corresponding to its minimum anisotropy constant K_1. The compositional dependence of the ratio λ_a/K_1 for the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ $(0.10 \leqslant x \leqslant 0.28)$ alloys is presented in Fig. 5. It can be seen that every curve presents a peak at $x=0.22$ when the magnetic field varies from 1 to 12 kOe. The piezomagnetic coefficient d_{33} of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$/epoxy composites was measured and it was found that the $d_{33\,max}$ for $x=0.22$ is the largest at relatively low fields.[18] These results also indicate that the $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy has a good low-field magnetostrictive property, which makes it a good candidate for magnetostriction applications.

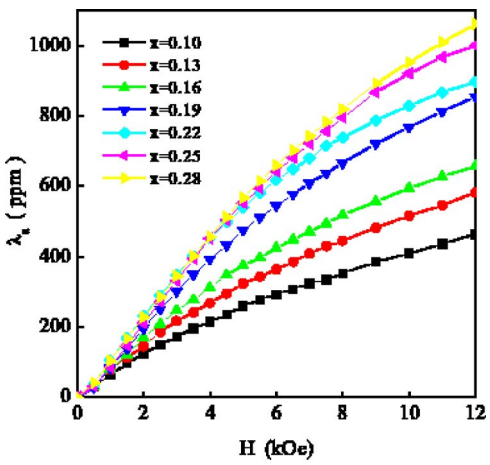

FIG. 4. (Color online) Magnetic field dependence of the magnetostriction λ_a $(=\lambda_\parallel-\lambda_\perp)$ for the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys.

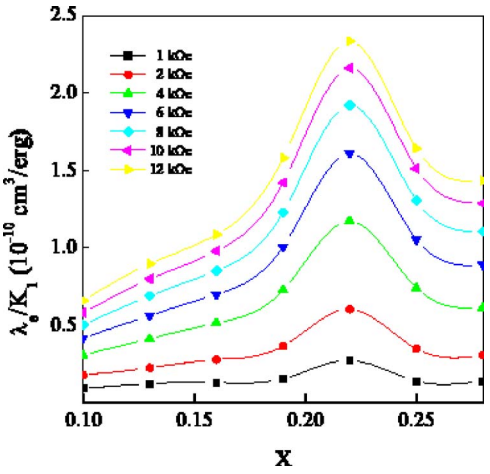

FIG. 5. (Color online) Compositional dependence of the ratio λ_a/K_1 for the $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloys.

The step-scanned (440) XRD line of the $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy, which was deducted of $K\alpha_2$ with a standard method, is represented in Fig. 6. The solid line is experimental data. The dash lines are fitted XRD lines, corresponding to the pseudocubic (440) and $(44\bar{0})$ line of the compound with a rhombohedral distortion caused by the magnetostriction. The (440) split line is consistent with its EMD along the ⟨111⟩ direction.[13,14] The dot line results to be the sum of the two fitted lines. The spontaneous magnetostriction λ_{111} can be obtained from the fitted XRD lines

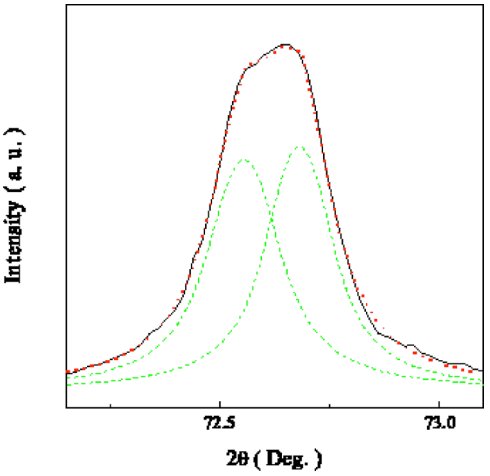

FIG. 6. (Color online) The (440) XRD line and the fitted lines of the $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy.

using the equation $\lambda_{111}=2(d_{440}-d_{44\bar{0}})/(d_{440}+d_{44\bar{0}})$, where d_{440} and $d_{44\bar{0}}$ denote the crystallographic plane distances of (440) and $(44\bar{0})$, respectively.[7,8] For this case, the value of the spontaneous magnetostriction λ_{111} was calculated to be 1500 ppm. Summarizing the results above, $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy has good magnetostrictive properties, that is, a large magnetostriction and low anisotropy at room temperature, and it contains 30 at. % low-cost Pr.

IV. CONCLUSION

In conclusion, the spin configuration and magnetostrictive properties of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ $(0.10 \leqslant x \leqslant 0.28)$ alloys have been investigated in detail. The Curie temperature T_C increases, while the spin reorientation temperature T_{SR} decreases as Tb content increases. The phase diagram for the spin configuration of $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ Laves phases has been constructed using the data of T_C, T_{SR}, and the results of EMD. The magnetocrystalline anisotropy at room temperature decreases as Tb content increases and reaches a minimum value at $x=0.22$. It is remarkable that, at the same composition, λ_a/K_1 also exhibits a peak at different magnetic fields. From these results and their analysis it can be concluded that the $Tb_{0.22}Dy_{0.48}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ alloy with $\lambda_{111} \approx 1500$ ppm results to be a very promising magnetostrictive material for technological applications.

ACKNOWLEDGMENTS

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