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Influence of Tb on easy magnetization direction and magnetostriction of $PrFe_{1.9}$ alloy*

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The crystal structures, magnetization, and spontaneous magnetostriction of ferromagnetic Laves phase $Pr_{1-x}Tb_xFe_{1.9}$ compounds are investigated in a temperature range between 5 K and 300 K. High resolution synchrotron x-ray diffraction (XRD) analysis shows that different proportions of Tb in $Pr_{1-x}Tb_xFe_{1.9}$ alloys can result in different easy magnetization directions (EMD) below 70 K, *i.e.*, [100] with $x = 0.0$, and [111] with $x \geq 0.1$. This indicates Tb substitution can lead the EMD to change from [100] to [111] with x rising from 0.0 up to 0.1. The Tb substitution for Pr reduces the saturation magnetization M_s and the magnetostriction to their minimum value when $x = 0.6$, but it can increase low-field ($0 \leq H \leq 9$ kOe, the unit 1 Oe = 79.5775 A·m^{-1}) magnetostriction when $x = 0.8$ and 1.0 at 5 K. This can be attributed to the larger magnetostriction of $PrFe_{1.9}$ than that of $TbFe_{1.9}$, as well as the decrease of the resulting anisotropy due to Tb substitution at low temperatures.

Keywords: Tb substitution, spin reorientation, magnetostriction, easy magnetization direction (EMD)

PACS: 75.50.Bb, 75.80.+q **DOI:** 10.1088/1674-1056/ab47f6

1. Introduction

The binary C15 cubic Laves phase RFe_2 (R = rare-earth elements) compounds are known to exhibit giant magnetostriction, which has been widely used as magnetostrictive materials in transducers, spintronics, information storage, sensors, and the design of novel electronic devices.[1–9] Especially, the low-cost light rare earth $PrFe_2$ alloy, which has a large calculated magnetostriction ($\lambda_{111} \sim 5600$ ppm at 0 K) according to single ion model theory, has received much attention in recent years.[10–14] However, this alloy shows high magnetocrystalline anisotropy due to the strong anisotropic 4f shells, and thus it possesses high switching fields at low temperatures.[15] In the past few years, much attention has been paid to developing the anisotropy compensating system similar to the well-known Terfenol-D, by combining two different RFe_2 terminals with opposite signs of anisotropy constant, *e.g.*, $Tb_{0.2}Dy_{0.8-x}Pr_x(Fe_{0.9}B_{0.1})_{1.93}$, $Tb_xDy_{1-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$, and $Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.9}B_{0.1})_2$ systems, *etc*.[11,16,17] Among them, $Pr_{1-x}Tb_xFe_{1.9}$ is a promising anisotropy compensating system, while many Tb alloys have favorable characteristics at low magnetic fields.[18–23] Since the easy magnetic direction (EMD) of $PrFe_2$ lies along [100], while the easy magnetic direction of $TbFe_2$ lies along [111] at low temperatures,[1,15] their magnetocrystalline anisotropy constants with different signs may be offset by each other.[24] In addition, $TbFe_2$ has large magnetostriction ($\lambda_{111} \sim 4400$ ppm at 0 K) and the same sign of the magnetostriction as $PrFe_2$, which might enlarge the low-field magnetostriction in the $Pr_{1-x}Tb_xFe_{1.9}$ system with appropriate Tb component. Furthermore, according to the x-ray diffraction studies on $PrFe_2$, its easy magnetic direction lies along [111] above its spin reorientation temperature T_{sr} (about 70 K), while it lies along [100] when $T < T_{sr}$.[15] This transition is considered as a morphotropic phase boundary (MPB) phenomenon in ferromagnets, and it has driven the design of materials with giant low-field magnetostriction.[25–28] Nevertheless, most of this meaningful research mainly focuses on the heavy rare-earth system. Further MPB study on the low-cost light rare-earth containing systems is still needed, for example, the $Pr_{1-x}Tb_xFe_{1.9}$ system, in which $PrFe_{1.9}$ and $TbFe_{1.9}$ are the two starting components with different crystal structures.

The objective of the presented work is to study the effect of the rare-earth (Tb) substitution for Pr on the structural symmetry, spin reorientation temperature, magnetization, and magnetostriction in the ferromagnetic $Pr_{1-x}Tb_xFe_{1.9}$ alloys.

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2. Experiment design

Ingots with $Pr_{1-x}Tb_xFe_{1.9}$ ($x = 0.0$, 0.05, 0.1, 0.2, 0.4, 0.6, 0.8, 1.0) stoichiometry were prepared by melting the high purity metals in a magneto-controlled arc furnace in an argon atmosphere.[12,23] The alloys were prepared from materials with the following purities: Pr and Tb (99.9 wt%), Fe (99.8 wt%). The ingots each with about 1 g were pressed into disks and wrapped with tantalum foils, and then loaded into a cylindrical graphite pipe heater. The assembly was pressed to 6 GPa by a hexahedral anvil press and heated to 1173 K for 30 min. Conventional x-ray diffraction (XRD) analysis with using Cu-$K\alpha$ radiation with a Rigaku D/Max-ga diffractometer was made. The samples for XRD were ground into powders to reduce the preferred orientation effect. The powder XRD patterns for $Pr_{1-x}Tb_xFe_{1.9}$ alloys with different Tb concentrations at room temperature were obtained and are shown in Fig. 1.

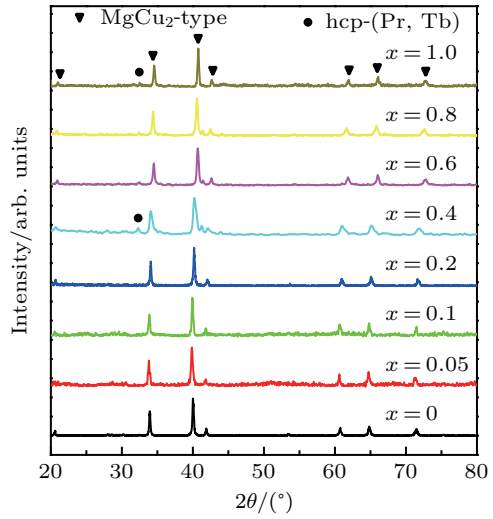


Fig. 1. XRD patterns for $Pr_{1-x}Tb_xFe_{1.9}$ ($x = 0.0$, 0.05, 0.1, 0.2, 0.4, 0.6, 0.8, 1.0) alloys.

It is found that all the polycrystalline alloys consist predominantly of cubic Laves phase with $MgCu_2$-type structure. It is generally believed that the cubic Laves phase could not be obtained under normal pressure in high-Pr alloys due to the large radius of Pr^{3+}. Therefore, the formation of the Laves phase in high-Pr alloy should be ascribed to the effect of the high-pressure annealing method.[12,23] There is a hint of impurity phase, *i.e.*, hcp-(Pr, Tb) coexisting in the alloys,[22,23] which is marked in the figure. High precision step scanning for $40.5° \leq 2\theta \leq 43°$ and $70.5° \leq 2\theta \leq 72.5°$ enabled the spontaneous magnetostriction-induced splitting of the {222} and {440} lines to be investigated, respectively. The XRD peaks were fitted by Jade 6.5 XRD analytical software, and the profiles of the {440} and {222} lines of $K\alpha_2$ were deducted with a standard method. Next, superconducting quantum interference device magnetometer (SQUID) was used to measure the magnetization curves of the samples. The temperature dependence of magnetization was also measured by using SQUID to determine the spin reorientation temperature T_{sr}. The magnetostriction was then measured by a standard strain-gauge technique in the parallel ($\lambda_{||}$) direction and perpendicular ($\lambda_{\perp}$) direction of magnetic field for each sample (a cuboid block with a size of about 6 mm× 4 mm× 1 mm for each sample), with the magnetic field supplied by "Quantum Design" (PPMS).

3. Results and discussion

The curves of magnetostriction $\lambda_{||}$ *versus* applied magnetic field H for various x values at 5 K are measured and shown in Fig. 2(a). It is found that the magnetostriction for each of all the alloys with different x values is far from saturation even when the magnetic field reaches a maximum value of 40 kOe, indicating the large magnetocrystalline anisotropy of the alloys at 5 K. The Tb substitution for Pr reduces $\lambda_{||}$ in the magnetic fields of 9 kOe$\leq H \leq$ 40 kOe. As shown in Fig. 2(b), the magnetostrictions $\lambda_{||}$ decreases appreciably with x increasing to a minimum value when $x \leq 0.6$, then increases with x increasing to 0.8, and finally decreases with x further increasing. This result is similar to that of $Tb_{1-x}Pr_x(Fe_{0.4}Co_{0.6})_{1.9}$ system, in which a minimum λ also appears when Tb composition content is about 0.5, due to the compensation for magnetic moment between $PrFe_{1.9}$ and $TbFe_{1.9}$ alloys.[29] The alloy with $x = 0.0$ possesses the largest $\lambda_{||}$ at 5 K in a range of magnetic field: 9 kOe$\leq H \leq$ 40 kOe, due to the larger magnetostriction of $PrFe_{1.9}$ ($\lambda_{111} \sim$ 5600 ppm at 0 K) than that of $TbFe_{1.9}$ ($\lambda_{111} \sim$ 4400 ppm at 0 K) at low temperatures.[1] However, Tb substitution to an appropriate extent is beneficial to increasing $\lambda_{||}$ in low magnetic fields. As shown in Fig. 2(c), the alloy with $x = 0.8$ and 1.0 each possess a larger value of magnetostrictions $\lambda_{||}$ than the other alloys in magnetic fields of $0 \leq H \leq 9$ kOe, indicating that Tb substitution can help to increase $\lambda_{||}$. This can be well explained by the fact that $\lambda_{||}$ of $PrFe_{1.9}$ is larger than that of $TbFe_{1.9}$ and the Tb substitution reduces the magnetocrystalline anisotropy.[1,23] For the RFe_2 compound with a cubic $MgCu_2$-type structure, the EMD lies along the [100] or [111] direction depending on whether the anisotropy constant K_1 is positive or negative, with K_2 and anisotropy constants neglected.[24] The EMD lies along the [100] direction for $PrFe_2$ and the direction [111] for $TbFe_2$ at low temperatures.[1,15] This indicates the different anisotropy indicators of the two alloys. Therefore, it can be expected that $Pr_{1-x}Tb_xFe_{1.9}$ compounds with appropriate compositions may have a small magnetic anisotropy but a large magnetostriction at low temperatures.

Figure 3 shows the temperature dependence of the magnetization (M) in a magnetic field of 5 kOe in a temperature range from15 K to 300 K. For the alloy with $x = 0.0$ shown in Fig. 3(a), the magnetization does not increase monotonically with temperature decreasing, but an extreme value appears at a spin reorientation temperature T_{sr} of about 70 K. This anomaly is mainly due to the occurrence of spin reorientation in the alloys,[15,19,26,30] which has been reported in our previous studies. From Fig. 3(b) to Fig. 3(f), we can see the monotonic increase of the magnetization (M) with temperature decreasing in the alloys with $0.1 \leq x \leq 1.0$, due to RE–TM and TM–TM interactions increasing as temperature decreases.[1] These phenomena indicate that the crystal structure might keep the same in the alloys with $0.1 \leq x \leq 1.0$ all over the temperatures investigated, and it will be discussed in Fig. 4 for details.

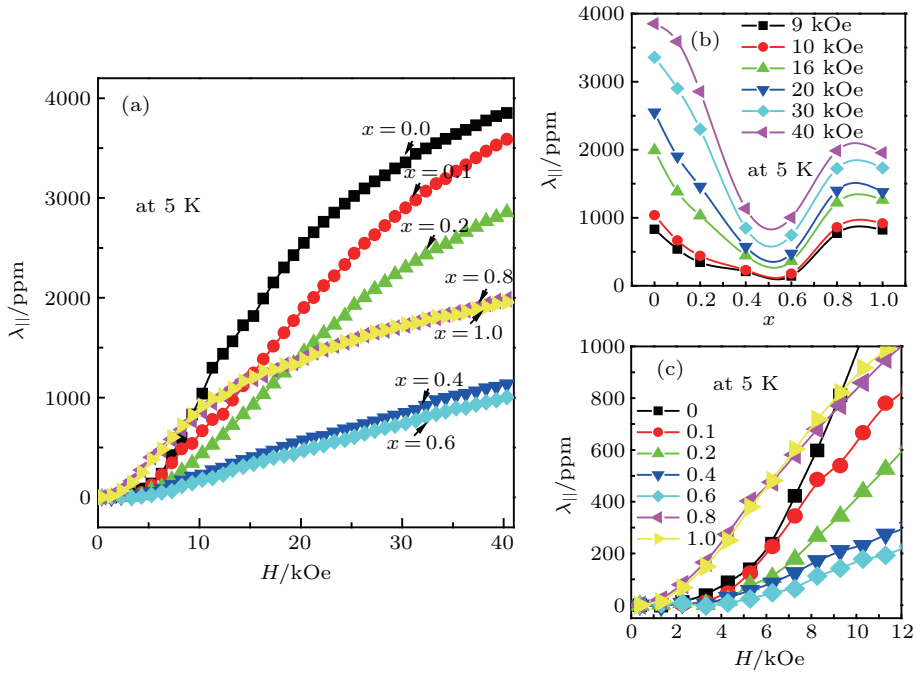


Fig. 2. (a) Plots of magnetostriction ($\lambda_{||}$) *versus* magnetic field of $Pr_{1-x}Tb_xFe_{1.9}$ alloys for various x values. (b) Plots of magnetostriction *versus* composition at different magnetic fields. (c) Magnified plots of panel (a) in magnetic field range of $0 \leq H \leq 12$ kOe.

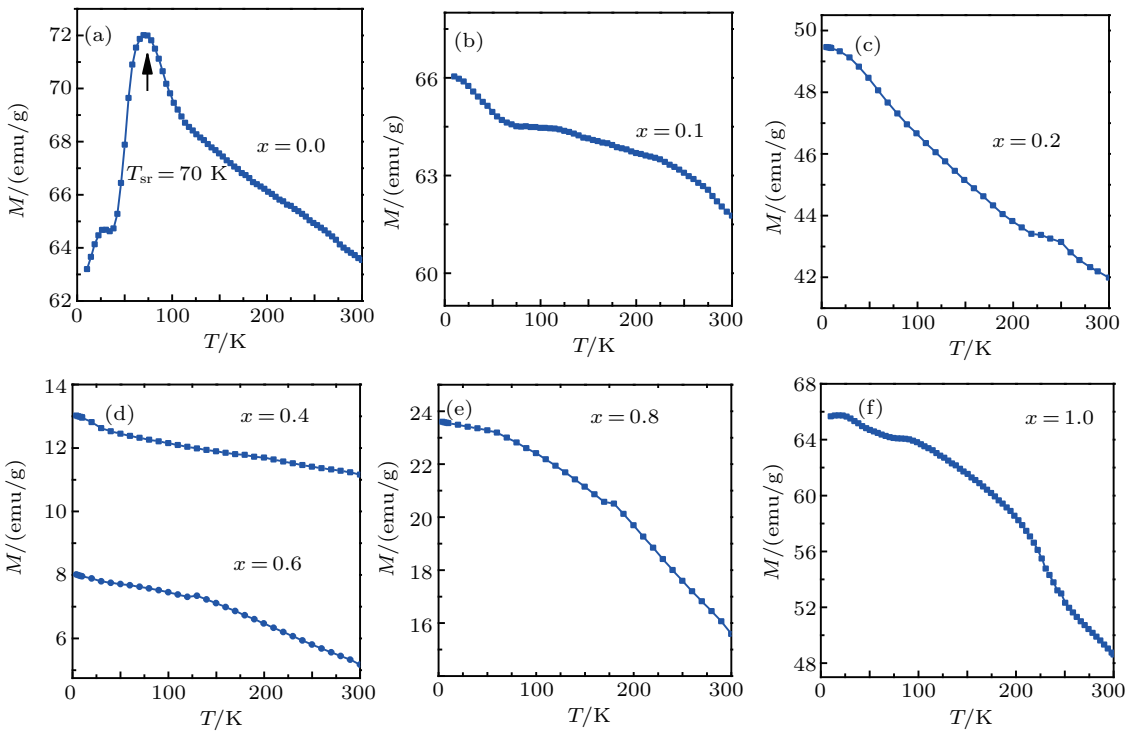


Fig. 3. Temperature dependence of magnetization (M) in the alloys with (a) $x = 0.0$, (b) $x = 0.1$, (c) $x = 0.2$, (d) $x = 0.4$, (e) $x = 0.8$, and (f) $x = 1.0$ in a magnetic field of 5 kOe.

The ferromagnetic transition can involve structural change.[19] In order to verify the possible change of EMD in such transitions, as well as the influence of Tb on the crystal structure in the alloys, a high-precision XRD step scanning is performed in the alloy with $x = 0.1$ under cooling condition, which is shown in Fig. 4(a). It can be seen from Fig. 4(a) that the sample demonstrates a rhombohedral symmetry in a temperature range between 15 K and 300 K. This is characterized by the {222} reflections doubly splitting with the intensity ratio of split peaks about 1:3 (corresponding to the pseudo-cubic (222) and ($\bar{2}22$) reflections), and double-splitting of {440} reflections with the intensity ratio of split peaks about 1:1 (corresponding to the pseudo-cubic (440) and ($44\bar{0}$) reflections), respectively.[2,15,18,19,25,31–33] This result indicates that the EMD of $Pr_{0.9}Tb_{0.1}Fe_{1.9}$ lies along the [111] direction at all of the temperatures investigated, which accords well with the magnetization result in Fig. 3(b). To study the influence of Tb substitution on the EMD, the XRD profiles of $TbFe_{1.9}$ and $PrFe_{1.9}$ are shown in Figs. 4(b) and 4(c), respectively. Figure 4(b) indicates that the $TbFe_{1.9}$ possesses a typical rhombohedral symmetry at all temperatures investigated, corresponding to the EMD lying along the [111] direction. On the other hand, figure 4(c) indicates that the $PrFe_{1.9}$ demonstrates a rhombohedral symmetry (the same as $TbFe_{1.9}$ and $Pr_{0.9}Tb_{0.1}Fe_{1.9}$) at 140 K. But at 15 K it demonstrates a tetragonal symmetry with the non-splitting of {222} reflections, and double-splitting of {440} reflections (corresponding to the pseudo-cubic (440) and (044) reflections).[2,25] Comparing with the magnetization (M) result in Fig. 3(a), the EMD of $PrFe_{1.9}$ lies along the [111] direction above 70 K, but along the [100] direction below 70 K. This indicates different types of EMD in $Pr_{1-x}Tb_xFe_{1.9}$ with different proportions of Tb below 70 K (*i.e.*, EMD along the [100] direction when $x = 0.0$, EMD along the [111] direction when $x \geq 0.1$). Then, we can come to the conclusion that in the temperature range from 15 K to 70 K, Tb substitution with $x \geq 0.1$ can lead the EMD to change from [100] to [111]. This result is also consistent with the room temperature result of $Tb_xDy_{1-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ system reported by Ren, in which the EMD changing from [100] to [111] occurs as Tb compensation increases from 0.15 to 0.25.[16] Meanwhile, this is also similar to the result of $Gd_{1-x}Tb_xFe_2$ system, in which the replacement of Gd with a small amount of Tb ($x = 0.1$) also changes the EMD from [100] to [111] at room temperature.[18]

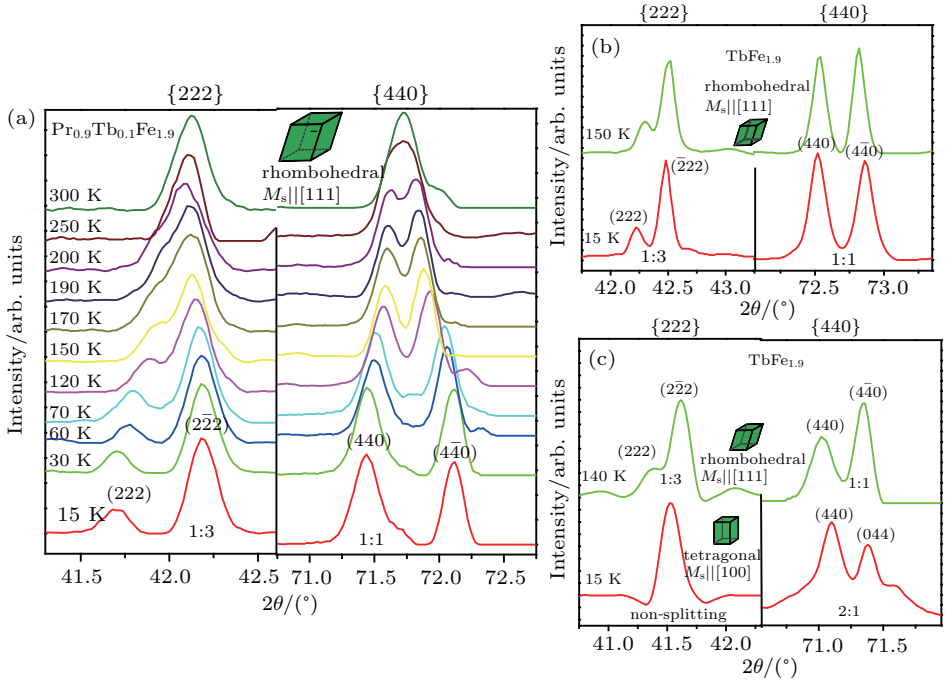


Fig. 4. Profiles of step-scanned {222} and {440} reflections in (a) $Pr_{0.9}Tb_{0.1}Fe_{1.9}$, (b) $PrFe_{1.9}$, and (c) $TbFe_{1.9}$, respectively.

To further investigate the effect of Tb substitution for Pr on the magnetizations and the magnetocrystalline anisotropy, the magnetization curves of $Pr_{1-x}Tb_xFe_{1.9}$ are measured at 5 K and 300 K, which are shown in Figs. 5(a) and5(b), respectively. The Tb substitution for Pr reduces the value of saturation magnetizations M_s when $x \geq 0.6$, but it is beneficial to reducing the magnetocrystalline anisotropy when $x = 0.8$ and 1.0 at 5 K. As shown in Fig. 5(a), the magnetization at 70 kOe decreases with Tb concentration increasing till $x = 0.6$ and then increases when $x \geq 0.6$. This is highly coincident with the magnetostriction result in Fig. 2(a), and is also similar to the result at room temperature, reported by Shi.[23] Since Fe couples ferromagnetically with Pr and couples antiferromagnetically with Tb respectively, the magnetic moment of

$Pr_{1-x}Tb_xFe_{1.9}$ can be described as $\mu_s = (1-x)\mu_{Tb} - 1.9\mu_{Fe} - x\mu_{Pr}$.[1,23] Therefore, the Tb substitution for Pr leads the saturation magnetization to decrease and subsequently increase. The magnetic field dependence of the normalized magnetization $M/M_{(70\ kOe)}$ at 5 K is presented in the inset of Fig. 5(a) to investigate the magnetocrystalline anisotropy of the alloys. It is found that in a magnetic field range of $0 \le H \le 4$ kOe, the alloys with $x = 0.8$ and 1.0 possess larger $M/M_{(70\ kOe)}$ value than the other alloys. This indicates that Tb substitution in $PrFe_{1.9}$ is a successful approach to reducing the magnetic anisotropy at 5 K. Then, the fast increase of the magnetostriction in low magnetic fields in the Tb-contained alloy shown in Fig. 2(c) can be well explained by the decrease of the magnetocrystalline anisotropy in the alloys due to Tb substitution. Furthermore, the minimum $M/M_{(70\ kOe)}$ value is found with $x = 0.6$, which indicates the largest magnetocrystalline anisotropy in the alloy in the whole range of magnetic fields. This result coincides well with the magnetostriction result in Fig. 2(a), in which a minimum $\lambda_{||}$ value also appears in the alloy with $x = 0.6$.

As for the magnetization result at 300 K, the magnetization decreases with Tb concentration increasing till $x = 0.6$ and increases when $x \ge 0.6$, which is similar to the result at 5 K. The alloys with $x = 0.0$ possess the maximum $M/M_{(70\ kOe)}$ value as shown in the inset of Fig. 5(b). This indicates that Tb substitution with $x \ge 0.2$ cannot help to reduce the magnetocrystalline anisotropy in $Pr_{1-x}Tb_xFe_{1.9}$ alloys, which can be ascribed to the magnetocrystalline anisotropy of $TbFe_{1.9}$ being larger than that of $PrFe_{1.9}$ at room temperature.[22,34] This result is unlike the result at 5 K shown in Fig. 5(a), in which Tb substitution reduces the magnetocrystalline anisotropy when $x = 0.8$ and 1.0. It has been reported that Tb substitution reduces the magnetocrystalline anisotropy in $Tb_xPr_{1-x}(Fe_{0.8}Co_{0.2})_{1.9}$ system when $x = 0.05$ at room temperature.[23] Therefore, a smaller magnetocrystalline anisotropy in the $Pr_{1-x}Tb_xFe_{1.9}$ system might require a lower Tb concentration for the anisotropy compensation,[22,23] which can be discussed in the future work.

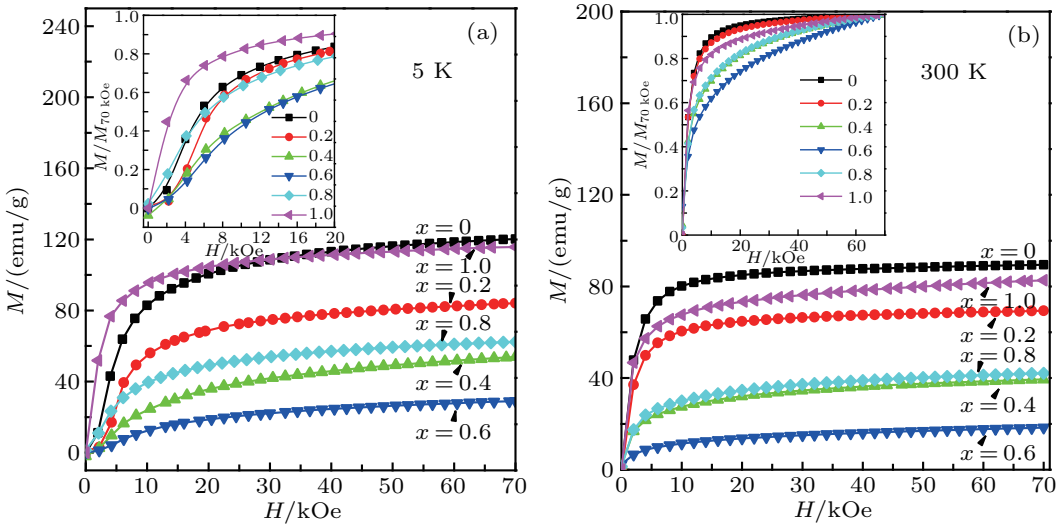


Fig. 5. Magnetic field dependence of magnetization (M) at (a) 5 K and (b) 300 K in polycrystalline $Pr_{1-x}Tb_xFe_{1.9}$ alloys for various x values. Inset shows the normalization magnetizations (M/M_{70kOe}) in alloys.

4. Conclusions

In conclusion, the substitution of Tb for Pr leads magnetostriction and saturation magnetization M_s to reach their corresponding minimum values when $x = 0.6$, but it also increases the value of the magnetostriction in a magnetic field range of $0 \le H \le 9$ kOe and reduces the magnetocrystalline anisotropy when $x = 0.8$ and $x = 1.0$ at 5 K. The easy magnetization direction (EMD) lies along the [100] direction in a temperature range between 15 K to 70 K when $x = 0.0$, but lies along the [111] direction when $x \ge 0.1$. This indicates that the Tb substitution with $x \ge 0.1$ can lead the EMD to change from [100] to [111].

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