# Low temperature magnetic and magnetostrictive properties in $Pr(Fe_{1-x}Co_x)_{1.9}$ cubic Laves alloys*

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(Received 28 August 2017; revised manuscript received 9 September 2017; published online 20 November 2017)

The structures, spin reorientations, magnetic, and magnetostrictive properties of the polycrystalline $Pr(Fe_{1-x}Co_x)_{1.9}$ ($x = 0$–1.0) cubic laves phase alloys between 5 K and 300 K are investigated. Large low-field magnetostrictions are observed at 5 K in the alloys with $x = 0.2$ and 0.4 due to the low magnetic anisotropies of these two alloys. A large negative magnetostriction of about −1130 ppm is found in $PrCo_{1.9}$ alloy at 5 K. The magnetizations of the alloys with $0 \leq x \leq 0.6$ decrease abnormally at the spin reorientation temperature $T_{sr}$, and an abnormity is detected in the alloy with $x = 1.0$ at its Curie temperature $T_c$ (45 K). The substitution of Fe by Co increases the value of $T_{sr}$ in the alloy with $x$ value increasing from 0.0 to 0.4, and then reduces the value of $T_{sr}$ with $x$ value further increasing to 0.6.

**Keywords:** magnetostriction, magnetization, easy magnetization direction (EMD)

**PACS:** 75.50.Bb, 75.80.+q **DOI:** 10.1088/1674-1056/26/12/127502

## 1. Introduction

According to theoretical calculations, the light rare earth Pr-based alloys should be potential alloys for magnetostrictive application, because of the low price of Pr and the theoretically predicted giant magnetostriction of $PrFe_2$ (5600 ppm) at 0 K.[1–10] However, their applications at low temperatures are somewhat limited due to their high magnetocrystalline anisotropy, which makes these materials hard to be saturated at low temperatures.[1–4] According to previous studies, the substitutions of Fe by the elements, such as Al, Mn, Ga, Co, Ni, Ti, etc., can help to reduce the magnetocrystalline anisotropy and improve the low-field magnetostrictive properties of $RFe_2$ alloy.[6–10] The substitution of Fe by Co increases the magnetostrictions of $Sm_{0.9}Pr_{0.1}(Fe_xCo_{1-x})_2$ alloys at room temperature,[5] and leads to the large magnetostrictions of $Pr(Fe_{0.8}Co_{0.2})_{1.9}$ and $Pr(Fe_{0.6}Co_{0.4})_{1.9}$ alloys in $Pr(Fe_{1-x}Co_x)_{1.9}$ system at room temperature,[3] etc. These researches indicate that the substitution of Fe by Co might be an ideal method to improve the magnetostrictive properties of $PrFe_2$ alloy. Moreover, previous studies showed that the easy magnetization direction (EMD) of $PrFe_2$ lies along the [100] direction at low temperatures and changes to the [111] direction at near 70 K,[4] while the EMD of $PrCo_2$ is along the [100] direction below its Curie temperature $T_c$ (about 49 K).[10] For $RFe_2$ 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, if $K_2$ and higher-order anisotropy constants are neglected.[11] The different EMDs of $PrFe_2$ and $PrCo_2$ at low temperatures indicate their different anisotropy signs. Therefore, it may be expected that $Pr(Fe_{1-x}Co_x)_{1.9}$ compounds with appropriate compositions may have a small magnetic anisotropy but, a large magnetostriction at low temperatures.

In this paper, the structural, magnetic and magnetostrictive properties of $Pr(Fe_{1-x}Co_x)_{1.9}$ laves compounds are investigated.

## 2. Experiment

Ingots with $Pr(Co_xFe_{1-x})_{1.9}$ ($x = 0$–1.0) were prepared by melting the high purity metals in a magneto-controlled arc furnace in an argon atmosphere.[3,12] The purities of constituents are 99.9% for Pr, 99.8% for Fe and 99.8% for Co, respectively. The ingots (about 1 g for each samples) were pressed into disks and each disk was wrapped with a tantalum foil, and then they were loaded into a graphite pipe heater with a cylindrical shape. The assembly was pressed to 6 GPa by a hexahedral anvil press and heated at 900 °C for 30 min. Conventional x-ray diffraction (XRD) analysis was carried out using Cu K$\alpha$ radiation with a Rigaku D/Max-ga diffractometer. The samples for XRD were ground into powders to reduce the preferred orientation effect. The linear magnetostriction in the parallel ($\lambda_{\parallel}$) direction and perpendicular ($\lambda_{\perp}$) direction of magnetic field for each sample (a rectangular block with a size of about 6 mm×4 mm×1 mm for each sample) was

*Project supported by the National Natural Science Foundation of China (Grant No. U1232210), the Science Foundation of Guangxi Zhuang Autonomous Region, China (Grant No. 2015GXNSFBA139020), and the Project of the Enhancement of the Basic Ability of Teachers of Guangxi Zhuang Autonomous Region, China (Grant No. KY2016YB068).

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measured by standard strain-gauge technique. The temperatures and magnetic-fields of the samples were controlled by the physical property measurement system (PPMS, Quantum design). The PPMS was also used to measure the initial magnetization curves and the temperature dependence of magnetization of the samples (a grain close to an ellipsoid with the aspect ratio $k \geq 2$ for each sample).

## 3. Results and discussion

Figure 1 shows the powder XRD patterns for $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys at room temperature. All of the $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys exhibit almost single $MgCu_2$-type cubic laves phase structure. It is known that the $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys with single cubic laves phase cannot be synthesized under ordinary pressure due to the large radius of $Pr^{3+}$ ion. Therefore, the successful syntheses of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys can be ascribed to the use of the high-pressure annealing method.[3,12]

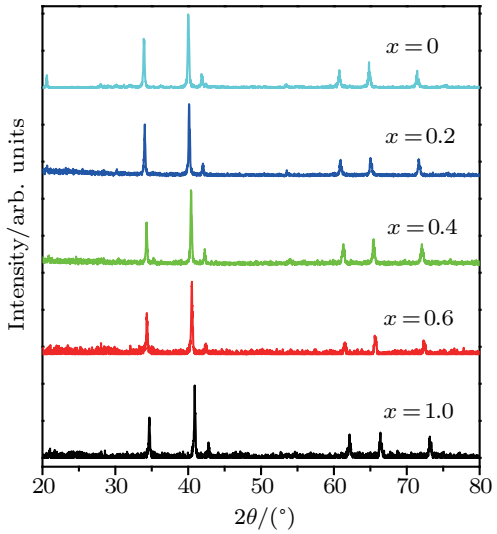



**Fig. 1.** (color online) XRD patterns for $Pr(Fe_{1-x}Co_x)_{1.9}$ ($x = 0$–1.0) alloys.

The field dependence of the magnetostriction of $Pr(Fe_{1-x}Co_x)_{1.9}$ compounds are measured at 5 K either parallel ($\lambda_{\parallel}$) or perpendicular ($\lambda_{\perp}$) to the applied fields up to 70 kOe (1 Oe = 79.5775 A·m$^{-1}$, which are shown in Figs. 2(a) and 2(b), respectively. For the polycrystalline compound, the magnetostriction $\lambda$ can be obtained by $\lambda = \lambda_{\parallel} - \lambda_{\perp}$. The values of $\lambda$ in $Pr(Fe_{1-x}Co_x)_{1.9}$ compounds versus external magnetic field at 5 K are shown in Fig. 2(c). For the alloys with $0 \leq x \leq 0.6$, the magnetostrictions decrease sharply from about 6700 ppm to about 800 ppm with $x$ increasing from 0.0 to 0.6, at a magnetic field of 70 kOe at 5 K. This indicates the substitution of Fe by Co reduces the magnetostriction at high magnetic field. Moreover, some particular features can be seen in the alloy with $x = 1.0$. The magnetostriction of $PrCo_{1.9}$ alloy is negative and initially goes to a negative maximum, then increases slightly with further increasing the magnetic field. This character is similar to the case of $Dy_{1-x}Pr_x(Fe_{0.35}Co_{0.55}B_{0.1})_2$ alloys[9] and $DyCo_2$ alloy.[13] Meanwhile, a large negative magnetostriction of about $-1130$ ppm is found in $PrCo_{1.9}$ alloy at 5 K at a magnetic field of 70 kOe. Large negative magnetostriction was also reported by previous study, the magnetostriction constant $\lambda_{100}$ of $PrCo_2$ alloy was predicted to be as large as about $-3400$ ppm at 0 K.[8] The large negative magnetostriction detected here in $PrCo_{1.9}$ alloy is similar to that of $NdCo_2$ alloy, in which the magnetostriction constant $\lambda_{100}$ is predicted to be as $-4000$ ppm at 0 K. Another highlight in Fig. 2(c) is that the substitution of Fe by Co improves the low-field magnetostrictive properties of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys. The inset of Fig. 2 shows the composition dependence of the magnetostriction of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys in a magnetic field range of $4 \leq H \leq 14$ kOe. It can be seen that the alloy with $x = 0.2$ has the largest magnetostriction at a magnetic field of 8 kOe, and the alloy with $x = 0.4$ has the largest magnetostriction at a magnetic field of 4 kOe. This result indicates that the appropriate substitution ($x = 0.2$, 0.4) of Fe by Co is beneficial to the increasing of the magnetostriction in low-magnetic field at 5 K although the magnetostriction decreases at high-magnetic field, which is similar to previously reported results of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys at room temperature.[3]

It is known that low magnetocrystalline anisotropy can lead to large low filed magnetostriction.[1–3] Therefore, to further study the magnetocrystalline anisotropies of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys at 5 K, their applied magnetic field dependence of the magnetization ($M$) at 5 K is measured as shown in Fig. 3(a). The magnetizations of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys decrease drastically with the increasing $x$ value at a high magnetic field of 70 kOe. This can be explained by the fact that the Co moment in $PrCo_2$ compound (0.85 $\mu_B$) is lower than the Fe moment (1.32 $\mu_B$) in $PrFe_2$ compound.[1,10] For $PrFe_{1.9}$ alloy, the magnetocrystalline anisotropy is quite large at low temperatures, which renders the alloy difficult to saturate even at 70 kOe. Magnetic anisotropy is a crucial property for a magnetostrictive material.[11] The magnetic field dependence of the normalized magnetization $M/M_{(70\ \mathrm{kOe})}$ at 5 K in a magnetic field range of $0 \leq H \leq 30$ kOe of $Pr(Fe_{1-x}Co_x)_{1.9}$ compounds presented in Fig. 3(b) shows the influence of Co substitution for Fe on the magnetic anisotropy. It is found from the figure that the alloy with $x = 1.0$ has the largest value of $M/M_{(70\ \mathrm{kOe})}$, which indicates that $PrCo_{1.9}$ has the smallest magnetocrystalline anisotropy in all of the alloys. As for the alloys with $0 \leq x \leq 0.6$, the value of $M/M_{(70\ \mathrm{kOe})}$ first increases to a maximum with $x$ increasing from 0.0 to 0.2, and then decreases with $x$ further increasing from 0.4 to 0.6. This indicates that the alloy with $x = 0.2$ has the smallest value of magnetocrystalline anisotropy in the alloys with $x \leq 0.6$. Then, the large low-field values of $\lambda$ in the alloys with $x = 0.2$, 0.4 in the inset of Fig. 2(c) can be well explained by the low magnetocrystalline anisotropies of these two alloys.

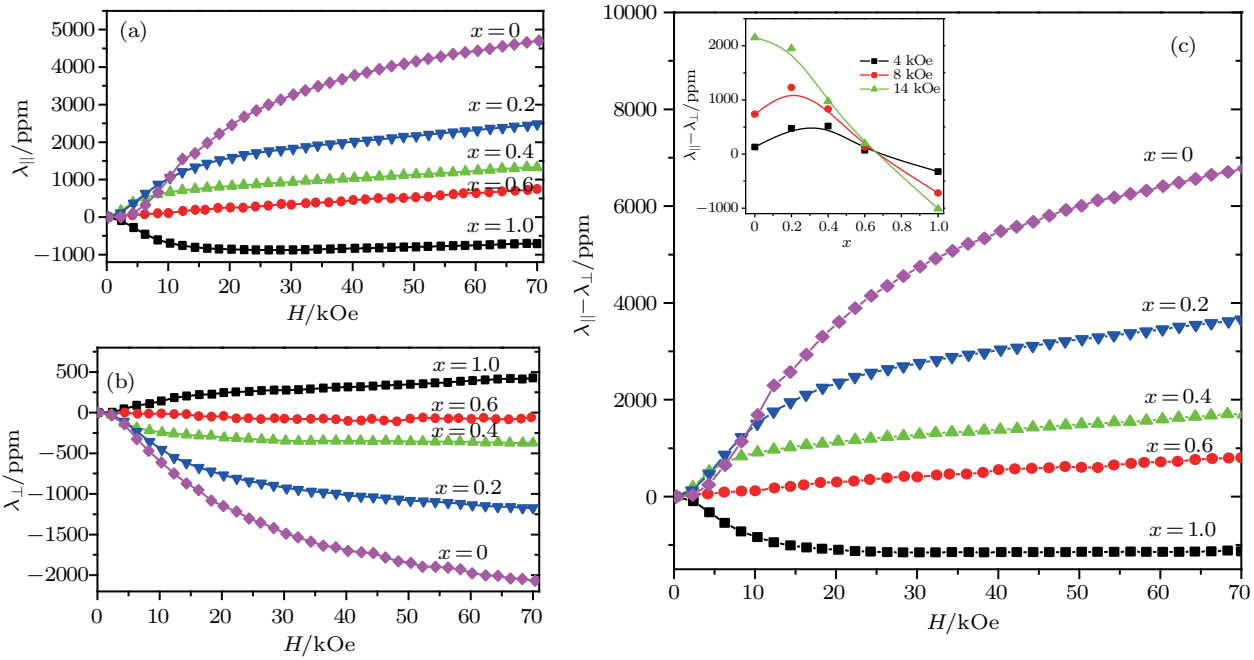



**Fig. 2.** (color online) Magnetic field dependence of the magnetostriction in the directions (a) parallel ($\lambda_{\parallel}$) and (b) perpendicular ($\lambda_{\perp}$) to the applied fields. (c) Magnetic field dependence of the magnetostriction ($\lambda = \lambda_{\parallel} - \lambda_{\perp}$) of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys. The inset shows the composition dependence of magnetostriction $\lambda$ in low magnetic field range (4 kOe $\leq H \leq$ 14 kOe).

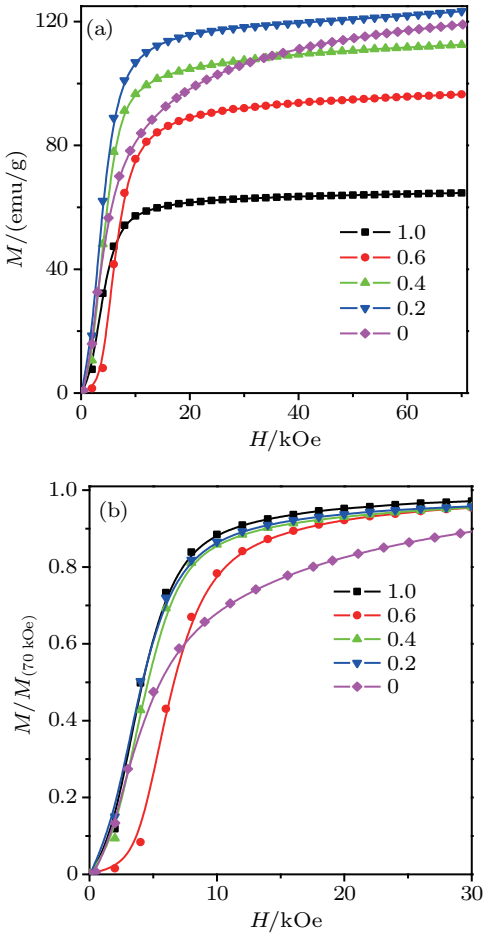



**Fig. 3.** (color online) (a) The initial magnetization curves of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys at 5 K, and (b) normalization magnetization $M/M_{(70\ \mathrm{kOe})}$ curves of $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys at 5 K.

The temperature dependence of the magnetization $M$ in $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys during cooling from 300 K to 10 K in a magnetic field of 5 kOe is measured as shown in Fig. 4. Several anomalies can be observed in the figure.

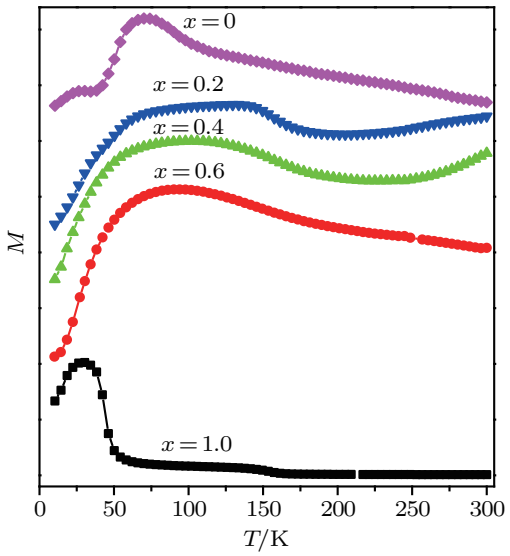



**Fig. 4.** (color online) The temperature dependence of the magnetization M in $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys ($H = 5$ kOe).

For the alloys with $0 \leq x \leq 0.6$, the magnetization $M$ values do not increase monotonically with temperature decreasing, but show some peak values at the spin reorientation temperature $T_{sr}$.[5,6,14–16] This peculiar feature is different from the feature of $TbFe_2$.[1] It has been reported that the Curie temperature $T_c$ of laves phase in $Pr(Fe_{1-x}Co_x)_{1.9}$ decreases from 510 K to about 400 K with $x$ increasing from 0.0 to 0.5.[3] Therefore, the abnormities of the magnetization $M$ at low tem-

peratures (below 150 K) detected here might not be caused by $T_c$. In consideration of that the EMD of $PrFe_{1.9}$ alloy changes from [111] to [100] between 30 K and 70 K,[4] therefore, the abnormal decreases of the magnetization in the alloys with $0 \leq x \leq 0.6$ are probably due to the change of the EMD in the alloys. The substitution of Fe by Co increases the value of the spin-reorientation temperature $T_{sr}$ (abnormal temperature in Fig. 4) with $x$ increasing from 0.0 to 0.4, and then reduces the value of $T_{sr}$ with the further increasing $x$ value to 0.6. This feature is similar to the result of $Pr_{0.5}Nd_{0.5}(Fe_{1-x}Co_x)_{1.9}$.[7] While for the alloy with $x = 1.0$, the magnetization sharply decreases to near-zero value above the Curie temperature $T_c$ (at around 45 K), which is in good agreement with the previously reported value (about 49 K).[10]

## 4. Conclusions

In this study, the substitution of Fe by Co increases the low-field magnetostrictions in the alloys with $x = 0.2$ and 0.4. The $PrCo_{1.9}$ alloy has a large negative magnetostriction of about $-1130$ ppm at 5 K. An abnormity is observed in the temperature dependence of the magnetization at $T_{sr}$ of the alloys with $0 \leq x \leq 0.6$, while it is observed at $T_c$ of $PrCo_{1.9}$.

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