# Magnetic properties and magnetostriction of $Pr_xNd_{1-x}Fe_{1.9}$ ($0 \leq x \leq 1.0$) alloys at low temperature*

Wang Yong(王 永), Tang Shao-Long(唐少龙)†, Li Yu-Long(李玉龙), Xie Ren(谢 忍), and Du You-Wei(都有为)

*Nanjing National Laboratory of Microstructures, Jiangsu Provincial Laboratory for Nanotechnology, and Department of Physics, Nanjing University, Nanjing* 210093, *China*

(Received 6 October 2012; revised manuscript received 24 October 2012)

The crystal structure, magnetic and magnetostrictive properties of high-pressure synthesized $Pr_xNd_{1-x}Fe_{1.9}$ ($0 \leq x \leq 1.0$) alloys were studied. The alloys exhibit single cubic Laves phase with $MgCu_2$-type structure. The initial magnetization curve reveals that $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ has a minimum magnetocrystalline anisotropy at 5 K. The magnetostriction curve at 5 K shows that $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ has a very good low-field magnetostrictive property, and the magnetostriction of the $Pr_xNd_{1-x}Fe_{1.9}$ alloy in high magnetic field is attributable mainly to Pr. The temperature dependence of the magnetostriction ($\lambda_{||}$) at the field of 5 kOe shows that the substitution of Nd reduces the $K_1$ remarkably, and the values of $\lambda_{||}$ of $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ and $Pr_{0.8}Nd_{0.2}Fe_{1.9}$ alloys are nearly five times larger than that of the $PrFe_{1.9}$ alloy below 50 K; the $\lambda_{||}$ of $Pr_{0.8}Nd_{0.2}Fe_{1.9}$ reaches up to 1082 ppm at 100 K, which makes it a potential candidate for application in this temperature range.

**Keywords:** magnetostriction, rare-earth compounds, cubic Laves phase, high-pressure annealing

**PACS:** 75.50.Bb, 75.80.+q **DOI:** 10.1088/1674-1056/22/3/037503

## 1. Introduction

In general, pseudobinary $RFe_2$ ($R$=rare-earth elements) compounds with cubic Laves phase exhibit giant magnetostriction values and large magnetocrystalline anisotropies at room temperature,[1–3] both of which have been investigated for many years. However, the large magnetocrystilline anisotropy $K_1$ leads to a small magnetostriction at a low field, which limits the practical applications of this kind of compounds. In order to reduce the magnetic anisotropy and improve the low-field magnetostriction, the $R_xR_{1-x}Fe_2$ anisotropy compensation system was proposed, in which the magnetostriction and the anisotropy of each $RFe_2$ alloy depend in a different way on the 4f electron wave function of $R$. Since then, the giant magnetostriction has been reported at room temperature in many binary compounds, like $Tb_xDy_{1-x}Fe_2$, $Tb_xHo_{1-x}Fe_2$, and $Tb_xNd_{1-x}Fe_{1.9}$.[3–14]

According to the single-ion model, the $PrFe_2$ alloy has a larger magnetostriction at low temperature,[1]and our recent investigation of $PrFe_{1.9}$ shows that it indeed has a huge magnetostriction at 5 K (3700 ppm in the direction parallel to a field of 50 kOe).[15] However, because of the large magnetocrystilline anisotropy, the $PrFe_{1.9}$ alloy does not show a good low-field magnetostriction. As is well known, the easy magnetization direction (EMD) of $PrFe_2$ changes from $\langle 100 \rangle$ to $\langle 111 \rangle$ as the temperature increases to room temperature,[16] while the EMD of $NdFe_2$ also changes from $\langle 110 \rangle$ to $\langle 100 \rangle$.[17] According to the single-ion model, $NdFe_2$ also has a large calculated magnetostriction of about 2000 ppm at 0 K.[1] In addition, a magnetostrictive compound with Pr or Nd should have good practical prospects considering their low price. Furthermore, our investigation of the Pr–Nd–Fe system at room temperature indicated that $Pr_xNd_{1-x}Fe_{1.9}$ might be a composition anisotropy compensation system.[18] Therefore, it is interesting to investigate the magnetostriction behavior of Pr–Nd–Fe systems at low temperature. In the present work, polycrystalline $Pr_xNd_{1-x}Fe_{1.9}$ ($0 \leq x \leq 1.0$) alloys with cubic Laves phase are prepared by using a high-pressure synthesis method, and the magnetostrictions of the alloys at low temperature are reported.

## 2. Method

The $Pr_xNd_{1-x}Fe_{1.9}$ ($x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0$) ingots were prepared by melting the constituent metals in a magneto-controlled arc furnace in a high-purity argon atmosphere. The purity of the constituents is 99.9% for Pr and Nd, and 99.99% for Fe. The as-cast ingot (about 1 g for each sample) was annealed at 6 GPa and 900 °C for 30 min. The conventional X-ray diffraction (XRD) analysis was implemented at room temperature with Cu $K_\alpha$ radiation with a Rigaku D/Max-gA diffractometer, and the samples for XRD were ground into powder to reduce the preferred orientation effect. Figure 1 shows the powder XRD patterns for the $Pr_xNd_{1-x}Fe_{1.9}$ alloys with different Pr concentrations at room temperature. All the samples show the single $MgCu_2$-type cubic Laves phase, which could be ascribed to high-pressure annealing.[15,18] The

*Project supported by the National Basic Research Program of China (Grant No. 2012CB932304), the National Natural Science Foundation of China (Grant Nos. 50831006 and 50771055), and the Priority Academic Program Development of Jiangsu Higher Education Institutions, China.

†Corresponding author. E-mail: tangsl@nju.edu.cn

© 2013 Chinese Physical Society and IOP Publishing Ltd 

Curie temperature was measured using a thermal gravitation analyzer with a vertical gradient magnetic field under the samples. A superconducting quantum interference device magnetometer (SQUID) was used to measure the magnetization curves of the samples. The magnetostriction was measured using the standard strain-gauge technique in parallel ($\lambda_{||}$) with the direction of the applied magnetic fields, and the magnetic field was supplied by a quantum design physical property measurement system (PPMS).

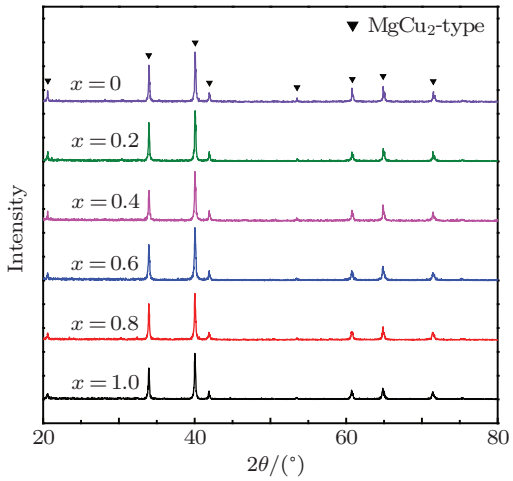



**Fig. 1.** (color online) XRD patterns of $Pr_xNd_{1-x}Fe_{1.9}$ with different Pr concentrations at room temperature.

## 3. Results and discussion

The magnetic-field dependence of the magnetization at 5 K for $Pr_xNd_{1-x}Fe_{1.9}$ was measured. The inset of Fig. 2 shows the initial magnetization curves of $Pr_xNd_{1-x}Fe_{1.9}$. It is evident that for alloys with high Pr concentrations, the magnetization values are not saturated up to 60 kOe. Figure 2 shows the $M/M_{max}$ versus $H$, where $M_{max}$ denotes the magnetization of the compound at 60 kOe. It can be seen that it is more difficult for the $PrFe_{1.9}$ alloy to be saturated than the $NdFe_{1.9}$ alloy,

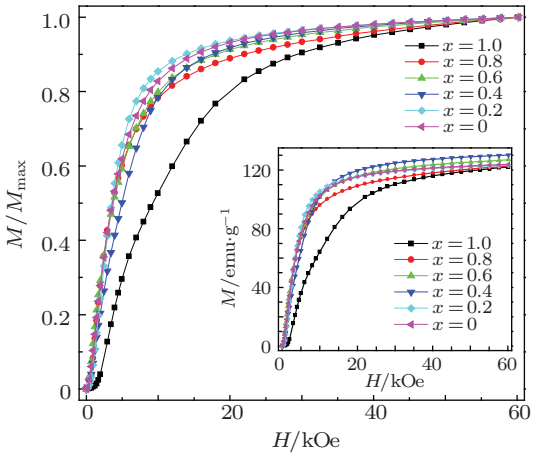



**Fig. 2.** (color online) The $M/M_{max}$ versus $H$ curves of $Pr_xNd_{1-x}Fe_{1.9}$ at 5 K, the variation of magnetization with the applied magnetic field is shown in the inset.

which indicates that $PrFe_{1.9}$ has a bigger magnetic anisotropy at this temperature. Among all the samples, $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ is the most easily saturated one, revealing that it has a lower magnetic anisotropy at 5 K in this ferromagnetic system. This result indicates that the moderate substitution of Nd for Pr indeed decreases the magnetic anisotropy at low temperature.

In order to confirm this conclusion, the magnetocrystalline anisotropy constant $K_1$ of the alloy at 5 K is determined by simulating the $M$–$H$ curve using the approximate saturation law[19]

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

with $b = (8/105)(K_1^2/\mu_0^2 M_s^2)$, where $M_s$ is the spontaneous magnetization, and $\chi_p$ is the paramagnetic susceptibility. Values of $K_1$ for all alloys are listed in Table 1. It can be seen that $K_1$ of $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ is the smallest one, and the substitution of Nd indeed decreases the magnetic anisotropy of $Pr_xNd_{1-x}Fe_{1.9}$ ($0 \le x \le 1.0$) alloys.

**Table 1.** Calculated magnetocrystalline anisotropy constant $K_1$ of $Pr_xNd_{1-x}Fe_{1.9}$ alloys at 5 K.

| $x$ | 1.0 | 0.8 | 0.6 | 0.4 | 0.2 | 0 |
|---|---|---|---|---|---|---|
| $K_1/10^5$ J·m$^{-3}$ | 26 | 16 | 13 | 10 | 6 | 7 |

The magnetostrictions of $Pr_xNd_{1-x}Fe_{1.9}$ alloys at 5 K are shown in Fig. 3, the measurement was performed in the direction parallel to the applied fields. It can be seen that at the largest field of 90 kOe, $\lambda_{||}$ increases from 1880 ppm ($x = 0$) to 4770 ppm ($x = 1.0$) with the increase of the Pr concentration. As far as we know, the data of 4770 ppm is the biggest experimental value for the $RFe_2$ compounds. It is also observed that $\lambda_{||}$'s of the alloys with low Pr concentrations ($x = 0$, 0.2, 0.4) are saturated at the field of 90 kOe, and the saturated magnetostriction $\lambda_{||}^S$ increases linearly with the increasing concentration of Pr, which can be described as

$$\lambda_{||}^S(x) = 1880 + \frac{870}{0.2}x. \tag{2}$$

According to Eq. (2), we can also evaluate the saturated magnetostrictions of alloys with high Pr concentrations ($x = 0.6$, 0.8, 1.0), the results are shown in Table 2. For isotropic polycrystalline materials, $\lambda_{||}^S$ can also be obtained by using the saturation relationship

$$\lambda_{||} = \lambda_{||}^S\left(1 - \frac{a}{H} - \frac{b}{H^2}\right), \tag{3}$$

where $a$ and $b$ are constants.[20,21] The saturated magnetostrictions calculated from this function are also summarized in Table 2. It is found from this table that the saturated magnetostrictions of each alloy given by the two different calculation methods are very close. This result reveals that $\lambda_{||}^S$ of the alloy with a high Pr concentration is also fit with Eq. (2). Therefore,

the increase of $\lambda_{||}^{S}$ of the $Pr_xNd_{1-x}Fe_{1.9}$ alloy obeys the linear relationship, which indicates that the magnetostriction of the $Pr_xNd_{1-x}Fe_{1.9}$ alloy at a high magnetic field mainly comes from the contribution of Pr.

**Table 2.** Calculated saturation magnetostriction values of $Pr_xNd_{1-x}Fe_{1.9}$ alloys at 5 K; $\lambda_{||\,a}^{S}$ and $\lambda_{||\,b}^{S}$ denote the calculated data obtained from Eqs. (2) and (3), respectively.

| $Pr_xNd_{1-x}Fe_{1.9}$ | $Pr_{0.6}Nd_{0.4}Fe_{1.9}$ | $Pr_{0.8}Nd_{0.2}Fe_{1.9}$ | $PrFe_{1.9}$ |
|---|---|---|---|
| $\lambda_{||\,a}^{S}$/ppm | 4490 | 5360 | 6230 |
| $\lambda_{||\,b}^{S}$/ppm | 4800 | 5500 | 5840 |

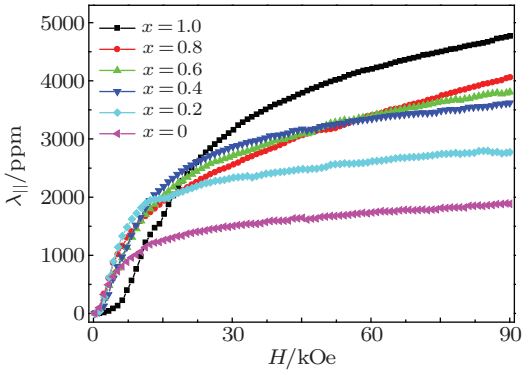



**Fig. 3.** (color online) Magnetic field dependence of $\lambda_{||}$ of $Pr_xNd_{1-x}Fe_{1.9}$ at 5 K.

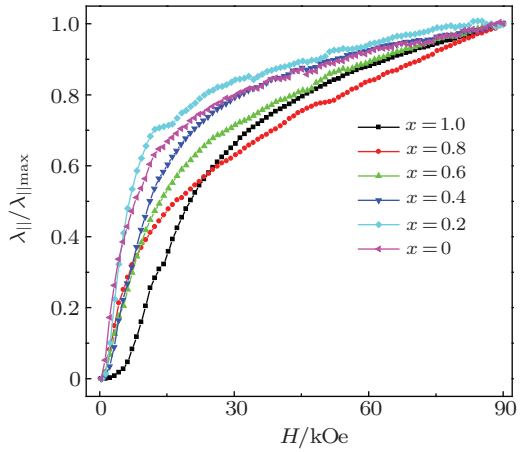



**Fig. 4.** (color online) The $\lambda_{||}/\lambda_{||\max}$ versus $H$ of $Pr_xNd_{1-x}Fe_{1.9}$ at 5 K.

From Fig. 3, it also can be seen that the alloys containing Nd have large low-field magnetostrictions, indicating that the substitution of Nd decreases the magnetic anisotropy of the alloy, which is consistent with the result in Fig. 2. It also can be seen that $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ possesses a better magnetostriction property at low fields, for example, $\lambda_{||}$ of $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ reaches up to 1140 ppm at the field of 5 kOe. Figure 4 shows the $\lambda_{||}/\lambda_{||\max}$ versus $H$, where $\lambda_{||\max}$ denotes the magnetostriction of the alloy at 90 kOe. It is evident from Fig. 4 that $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ is most easily saturated, suggesting that it has a lower magnetic anisotropy at 5 K. As we have mentioned above, the EMD of $PrFe_2$ is along the $\langle 100 \rangle$ direction at 5 K,[16] while that of $NdFe_2$ is along the $\langle 110 \rangle$ direction.[17] The $x = 0.2$ might be near the anisotropy compensation point of the Pr–Nd–Fe system at 5 K, which makes it have a smaller value of magnetic anisotropy and a better low-field magnetostrictive property.

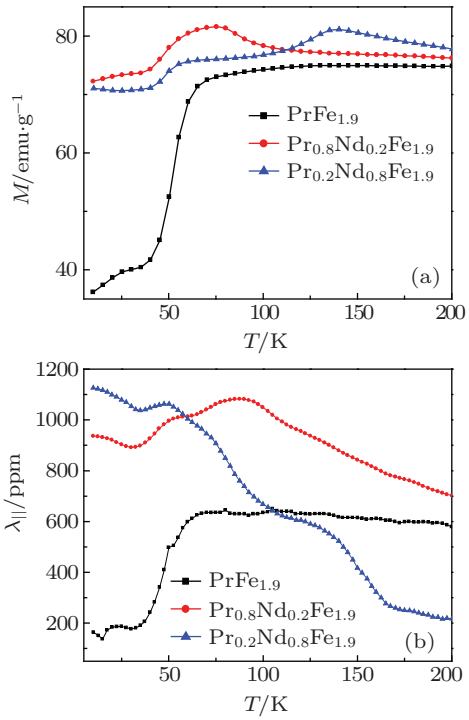



**Fig. 5.** (color online) Temperature dependences of (a) $M$ and (b) $\lambda_{||}$ of $PrFe_{1.9}$, $Pr_{0.8}Nd_{0.2}Fe_{1.9}$, and $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ in a magnetic field of 5 kOe.

Figure 5 shows the temperature-dependent magnetization $M$ and the magnetostriction $\lambda_{||}$ of $PrFe_{1.9}$, $Pr_{0.8}Nd_{0.2}Fe_{1.9}$, and $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ in the applied field of 5 kOe. From the $M$ vs. $T$ curve in Fig. 5(a), it is obvious that these alloys all have magnetic transitions as the temperature increases. It is known that the spontaneous magnetization will lead to a crystal-structural distortion of the alloy with the cubic $MgCu_2$-type Laves structure when the magnetization direction changes. Meanwhile, different ferromagnetic phases are coupled with different crystalline phases.[22,23] It can be seen in Fig. 5(a) that the magnetization state of $PrFe_{1.9}$ changes significantly, which is due to the change of the EMD as the temperature increases, and similar changes can be observed in the $\lambda_{||}$ vs. $T$ curve in Fig. 5(b). According to the results reported by Yang *et al.*, if the EMD lies along $\langle 111 \rangle$, a rhombohedral (R) distortion will occur; if the EMD lies along $\langle 100 \rangle$, the compound will have a tetragonal (T) crystal symmetry. Therefore, we suggest that the crystal structure change simultaneously occurs at the magnetic transition temperature and attribute the change of the magnetostriction to the different crystal structure distortion of the $PrFe_{1.9}$ alloy. As we have mentioned, the EMD

of $NdFe_2$ will change from $\langle 110 \rangle$ to $\langle 100 \rangle$ as the temperature increases;[17] hence, the substitution of Nd should have an influence on the EMD of the $Pr_xNd_{1-x}Fe_{1.9}$ alloys and should change the $\lambda_{||}$ of these alloys. It can be seen in Fig. 5(b) that the magnetostriction of the alloy with Nd changes at the magnetic transition temperature. It can also be seen that the $\lambda_{||}$'s of $Pr_{0.8}Nd_{0.2}Fe_{1.9}$ and $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ below 50 K are five times larger than that of $PrFe_{1.9}$, and that $\lambda_{||}$ of $Pr_{0.8}Nd_{0.2}Fe_{1.9}$ is also obviously bigger than that of $PrFe_{1.9}$ in the temperature range of 50–200 K. This result indicates that the substitution of Nd effectively reduces the magnetocrystalline anisotropy and improves the low-field magnetostriction in a large range of temperatures. Furthermore, the $\lambda_{||}$ of $Pr_{0.8}Nd_{0.2}Fe_{1.9}$ reaches up to 1082 ppm at 100 K, which makes it a candidate for practical applications around this temperature.

## 4. Conclusion

The initial magnetization curve of $Pr_xNd_{1-x}Fe_{1.9}$ at 5 K indicates that the magnetocrystalline anisotropy constant $K_1$ of the $PrFe_{1.9}$ alloy is bigger than that of the $NdFe_{1.9}$ alloy, and the $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ alloy shows a minimum value of $K_1$, which indicates that the substitution of Nd indeed decreases the $K_1$ of these alloys at low temperatures. The magnetostriction curve at 5 K shows that the $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ alloy has a better low field magnetostrictive property, and the magnetostriction of the $Pr_xNd_{1-x}Fe_{1.9}$ alloy at a high magnetic field is attributable mainly to Pr. The temperature dependence of the magnetostriction $\lambda_{||}$ of the $Pr_xNd_{1-x}Fe_{1.9}$ alloy at the field of 5 kOe shows that a moderate substitution of Nd reduces the magnetic anisotropy of the alloy remarkably, and the magnetostrictions of $Pr_{0.2}Nd_{0.8}Fe_{1.9}$ and $Pr_{0.8}Nd_{0.2}Fe_{1.9}$ are five times as large as that of $PrFe_{1.9}$ at low temperatures (below 50 K).

## References


[1] Clark A E 1980 *Ferromagnetic Materials* Vol. **1** (Amsterdam: Northholland) pp. 531–589

[2] Clark A E and deSavage B F 1966 *J. Appl. Phys.* **37** 1324

[3] Koon N C, Williams C M and Das B N 1991 *J. Magn. Magn. Mater.* **100** 173

[4] Clark A E, Abbundi R and Gillmor W R 1978 *IEEE Trans. Magn.* **14** 542

[5] Busbridge S C and Piercy A R 1993 *J. Appl. Phys.* **73** 5354

[6] Duenas T A and Carman G P 2000 *J. Appl. Phys.* **87** 4696

[7] Ren W J, Zhang Z D, Zhao X G, Liu W and Geng D Y 2004 *Appl. Phys. Lett.* **84** 562

[8] Hari Babu V, Markandeyulu G and Subrahmanyam A 2007 *Appl. Phys. Lett.* **90** 252513

[9] Jammalamadaka S N, Markandeyulu G and Balasubramaniam K 2008 *Appl. Phys. Lett.* **92** 044102

[10] Liu J J, Ren W J, Zhang Z D, Li D, Li J, Zhao X G, Liu W and Or S W 2006 *J. Appl. Phys.* **100** 023904

[11] Zeng D C and Zhang H 2010 *Acta Phys. Sin.* **59** 2808 (in Chinese)

[12] Zheng X P, Li F S, Fan D W, Zhang P F and Hao Y 2007 *Acta Phys. Sin.* **56** 535 (in Chinese)

[13] Zheng X P, Li F S, Fan D W, Zhang P F and Hao Y 2007 *Acta Phys. Sin.* **56** 535 (in Chinese)

[14] Cai N, Zhai J Y, Shi Z, Lin Y H and Nan C W 2004 *Chin. Phys.* **13** 1348

[15] Wang Y, Zhai L, Shi Y G, Tang S L and Du Y W 2010 *J. Appl. Phys.* **107** 103902

[16] Shimotomai M, Miyake H and Doyama M 1980 *J. Phys. F: Met. Phys.* **10** 707

[17] Meyer C, Hartmann-Boutron F, Gros Y, Bertheir Y and Buevoz J L 1981 *J. Phys.* **42** 605

[18] Shi Y G, Zhai L, Tang S L and Chen R J 2011 *J. Appl. Phys.* **109** 07A915

[19] Chikazumi S 1997 *Physics of Ferromagnetism* (2nd edn.) (New York: Oxford University Press) p. 503

[20] Liu J J, Wang R, Ying H Y, Liu X C and Du J 2011 *J. Appl. Phys.* **110** 073915

[21] Guo H Q, Gong H Y, Yang H Y, Li Y F, Yang L Y, Shen B G and Li R Q 1996 *Phys. Rev. B* **54** 4107

[22] Yang S, Bao H X, Zhou C, Wang Y, Ren X B, Matsushita Y, Katsuya Y, Tanaka M, Kobayashi K, Song X P and Gao J R 2010 *Phys. Rev. Lett.* **104** 197201

[23] Yang S and Ren X B 2008 *Phys. Rev. B* **77** 014407