Dy substitution effect on the temperature dependences of magnetostriction in $Pr_{1-x}Dy_xFe_{1.9}$ alloys*

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The temperature dependences of magnetostriction in $Pr_{1-x}Dy_xFe_{1.9}$ ($0 \le x \le 1.0$) alloys between 5 K and 300 K were investigated. An unusual decrease of magnetostriction with temperature decreasing was found in Pr-rich alloys ($0 \le x \le 0.2$), due to the change of the easy magnetization direction (EMD). Dy substitution reduces the magnetostriction in high-magnetic field (10 kOe $\le H \le$ 90 kOe) at 5 K, while a small amount of Dy substitution ($x = 0.05$) is beneficial to increasing the magnetostriction in low-magnetic field between 10 K and 50 K. This makes the alloys a potential candidate for low temperature applications.

Keywords: Dy substitution, magnetostriction, easy magnetization direction

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1. Introduction

The polycrystalline $PrFe_2$ alloy is of significant technological interest due to its giant magnetostriction (5600 ppm) at 0 K and the cheaper price of Pr than heavy rare-earth elements.[1,2] Recently, we reported that the magnetostriction λ_{111} of $PrFe_{1.9}$ alloy is as large as 6700 ppm at 70 K.[3] However, its application is somewhat limited, because $PrFe_{1.9}$ possesses large magnetocrystalline anisotropy below 100 K, which makes it difficult for the material to get saturated at low temperature.[3] Therefore, much attention should be paid to the preparation of $RR'Fe_2$ alloys or the substitution of Fe with the other metal element in $R'Fe_2$ alloys, for the purpose of developing new magnetostrictive materials in low temperature applications.[4–10] Among these elements, Dy is regarded as being ideal. Ren *et al.* reported that the anisotropies of $PrFe_2$ and $DyFe_2$ can compensate each other in the $Dy_{1-x}Pr_xFe_2$ system, as proved phenomenologically by a single-ion approach.[4,5] They found that the anisotropy of $Dy_{1-x}Pr_x(Fe_{0.9}B_{0.1})_{1.93}$ alloys decreases with the increase of x when $0 \le x \le 0.3$,[4] and the saturation magnetization M_s at 5 K or 295 K for $Dy_{1-x}Pr_x(Fe_{0.35}Co_{0.55}B_{0.1})_2$ alloys decreases to a minimum, then increases with the increase of the Pr content.[5] This meaningful work indicates that Dy substitution can help to decrease the magnetocrystalline anisotropy of $PrFe_{1.9}$ alloy, and might help to increase the low-field magnetostrictive properties at low temperature. Furthermore, Clark *et al.* found that the magnetostriction of $DyFe_2$ shows some abnormal features at low temperature (0 K$\le x \le$ 300 K).[1,11] And the investigation on step-scanned {440} and {222} x-ray diffraction (XRD) reflections of $PrFe_{1.9}$ alloy indicated that the easy magnetization direction (EMD) of the alloy changes from [111] to [100] with temperature decreasing.[3] The change of the EMD leads to the change of the magnetostriction in $PrFe_{1.9}$ alloy at some temperatures. Therefore, some particular features might be expected in the magnetostrictive property of $Pr_{1-x}Dy_xFe_{1.9}$ alloys, due to the particular magnetostrictive properties in $PrFe_{1.9}$ and $DyFe_2$ alloys.

In this paper, the temperature dependences of magnetostriction in the polycrystalline $Pr_{1-x}Dy_xFe_{1.9}$ ($0 \le x \le 1.0$) alloys between 5 K and 300 K were investigated in detail.

2. Experimental

The samples were prepared by the arc-melting and subsequent high pressure annealing.[12,13] Conventional XRD powders analysis was carried out using Cu $K\alpha$ radiation with a Rigaku D/Max-gA diffractometer. The powder XRD patterns for $Pr_{1-x}Dy_xFe_{1.9}$ alloys with different Dy concentrations at room temperature are shown Fig. 1. A single $MgCu_2$-type cubic Laves phase in all the samples investigated is confirmed, which could be ascribed to the high-pressure annealing method. The magnetostriction ($\lambda_{||}$) was measured by standard strain-gauge technique in the parallel direction of magnetic fields for each sample (a rectangular block with a size of about 6 mm×4 mm×1 mm for each sample), the magnetic field was supplied by a physical property measurement system (PPMS). A superconducting quantum interference device magnetometer (SQUID) was used to measure the magnetization curves of the samples.

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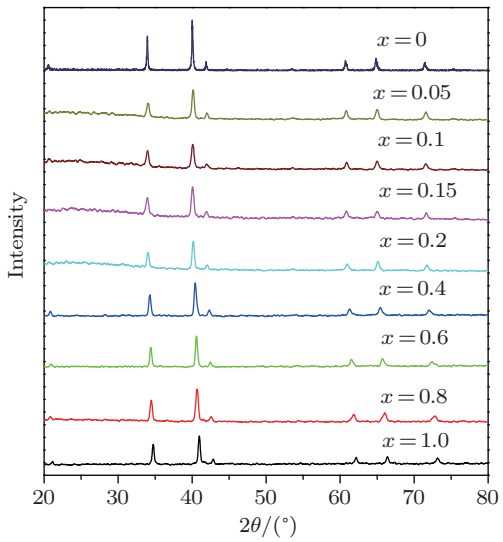


Fig. 1. (color online) XRD patterns for $Pr_{1-x}Dy_xFe_{1.9}$.

3. Results and discussion

Figure 2 shows the magnetostrictions ($\lambda_{||}$) as a function of the applied field at 5 K. It is found that the magnetostrictions of all the alloys with different x are far from saturation with the maximum magnetic field of 90 kOe. This indicates that the magnetocrystalline anisotropy of the alloys is quite large at 5 K. The slopes of the magnetostrictions curves in the alloys with $0 \leq x \leq 0.2$ are larger than those of the alloys with $0.2 \leq x \leq 1.0$. This indicates that the magnetocrystalline anisotropy of the alloys with $0 \leq x \leq 0.2$ is larger than that of the alloys with $0.2 \leq x \leq 1.0$ at 5 K. This tendency is similar to the results of the $Dy_{1-x}Pr_x(Fe_{0.9}B_{0.1})_{1.93}$ system and $Dy_{1-x}Pr_xFe_{1.9}$ system at room temperature.[4,14] In high-magnetic field ($10 \leq H \leq 90$ kOe), Dy substitution reduces the magnetostriction. The magnetostriction decreases sharply from 5000 ppm ($x = 0.0$) to 320 ppm ($x = 1.0$) in a magnetic field of 90 kOe. This might be due to that the magnetostriction of $PrFe_{1.9}$ is larger than $DyFe_{1.9}$.[1,2] However, in low magnetic field ($0 \leq H < 10$ kOe), Dy substitution yields an increased magnetostriction, which is shown in the inset of Fig. 2. It shows that the alloy with $x = 0.05$ has larger magnetostriction than the other alloys in the magnetic field of $0 \leq H < 10$ kOe, which indicates that a small amount of Dy substitution is beneficial to increase the magnetostriction at low temperature and in low magnetic field. This result is similar to the result reported by Ren.[5] They found in $Dy_{1-x}Pr_x(Fe_{0.35}Co_{0.55}B_{0.1})_2$ system that, at room temperature, $\lambda_{||}$ increases with the increase of Dy content when $0.8 \leq x \leq 1.0$, and $Dy_{0.2}Pr_{0.82}(Fe_{0.35}Co_{0.55}B_{0.1})_2$ possesses the largest magnetostriction.

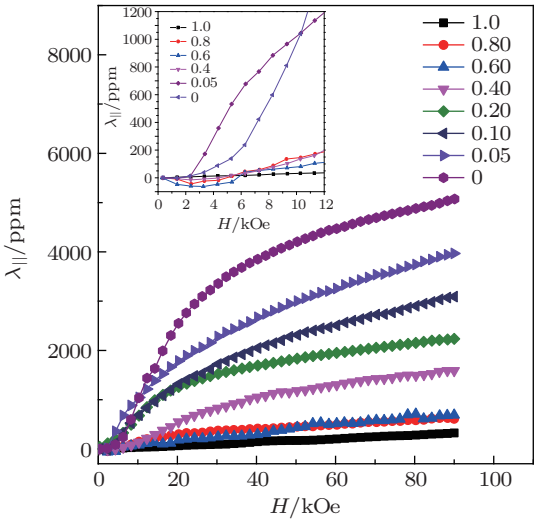


Fig. 2. (color online) The magnetostriction ($\lambda_{||}$) of $Pr_{1-x}Dy_xFe_{1.9}$ as a function of the applied field at 5 K. The inset shows the magnetostriction in the magnetic field of $0 \leq H \leq 1.2$ kOe.

The temperature dependences of magnetostriction ($\lambda_{||}$) during cooling from 300 K to 10 K in a magnetic field of 50 kOe were measured, which are shown in Fig. 3(a). Striking dissimilarity is found among these alloys. For Dy-rich alloys ($0.4 \leq x \leq 1.0$), the magnetostriction shows some peculiar features similar to that of $DyFe_2$, which are shown in Fig. 3(b). As seen in Fig. 3(b), as temperature decreases, the magnetostriction first decreases with the decrease of temperature to some temperature (at near 80 K for the sample $x = 1.0$), and then even changes sign with the further temperature decreasing. The temperature, at which the magnetostriction changes sign is near 80 K, 105 K, 185 K, and 150 K for $x = 1.0$, $x = 0.8$, $x = 0.6$, and $x = 0.4$, respectively. This particular feature was first reported by Clark in $DyFe_2$ alloy, and cannot fit a simple single-ion expression.[1,11] Therefore, the peculiar feature in Dy-rich alloys can be attributed mainly to the small and complex nature of λ_{100} of $DyFe_2$.[1,11,15] Recently, Bowden theoretically pointed out that this abnormality is due to the competition between $n = 2$, 4, and 6 crystal field distortions in $DyFe_2$ alloy, of which temperature dependences can be accurately modeled using the Callen and Callen model.[15] While for the Pr-rich alloys ($0 \leq x \leq 0.2$), the magnetostriction of the alloys has another unusual kind of feature, as shown in Fig. 3(a). It was found from Fig. 3(a) that, as the temperature decreases, the magnetostriction first increases to some temperature (at near 50 K for the sample $x = 1.0$), and then unusually decreases with the further temperature increasing. This peculiar feature is not like the feature of $TbFe_2$.[1] Recent study on the step-scanned {440} and {222} XRD reflections of $PrFe_{1.9}$ indicated that the EMD of the alloy changes from [111] to [100] with temperature decreasing between 30 K and 70 K. Therefore, we consider the abnormal decrease of the magnetostriction in Pr-rich alloys is probably due to the change of the EMD in the alloys.[3] For $PrFe_{1.9}$ alloy, the change of the EMD occurs between 70 K and 300 K, and for $Pr_{0.95}Dy_{0.05}Fe_{1.9}$ alloy, the change of the EMD might occur between 35 K and 120 K. This indicates that the increase of Dy

increases the spinreorientation temperature T_{SR} of the alloys. This result is similar to the results of $Tb_xDy_{1-x}(Fe_{0.8}Co_{0.2})_2$ and $Tb_xDy_{0.7-x}Pr_{0.3}(Fe_{0.9}B_{0.1})_{1.93}$ system reported by Liu *et al.*[16,17]

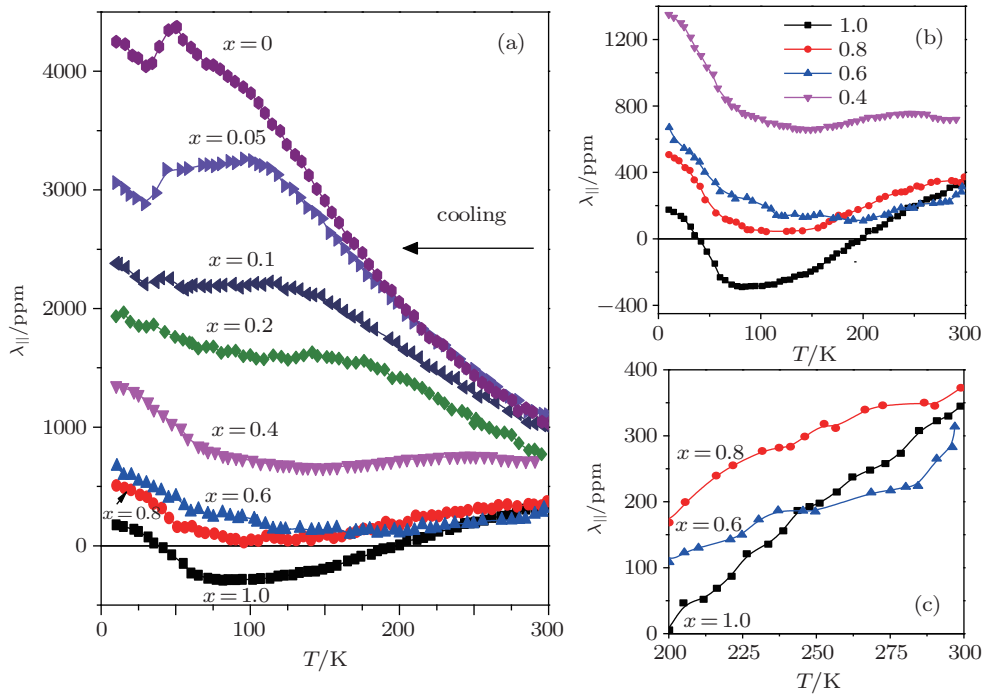


Fig. 3. (color online) (a) The temperature dependences of magnetostriction ($\lambda_{||}$) in $Pr_{1-x}Dy_xFe_{1.9}$ ($0 \le x \le 1.0$) alloys ($H = 50$ kOe). (b) The magnetostriction ($\lambda_{||}$) of the alloys with $0.4 \le x \le 1.0$ ($H = 50$ kOe). (c) The magnetostriction ($\lambda_{||}$) of the alloys with $0.6 \le x \le 1.0$ between 200 K and 300 K ($H = 50$ kOe).

Furthermore, it is found that Dy substitution for Pr with $x = 0.6$ yields a minimum $\lambda_{||}$ between 250 K and 300 K. As shown in Fig. 3(c), the magnetostriction of the sample with $x = 0.6$ increases from 184 ppm at 250 K to 313 ppm at 300 K, which is even smaller than the 197 ppm at 250 K to 344 ppm at 300 K of the sample with $x = 0.0$. This result agrees well with the result reported by Shi *et al.*,[14] in which the magnetostriction of $Dy_{0.6}Pr_{0.4}Fe_{1.9}$ also exhibits a minimum at room temperature. According to Clark, the magnetic moment of $Pr_{1-x}Dy_xFe_{1.9}$ can be described as: $\mu_s = x\mu_{Dy} - 1.9\mu_{Fe} - (1-x)\mu_{Pr}$.[1,5,14] And the decrease in Curie temperature with the increase in the atomic number is directly reflected in the room temperature value of the rare-earth sublattice magnetization.[1,4] Therefore, the magnetic moment compensation point at $x = 0.6$ between 250 K and 300 K can be well explained. The atomic number of Pr^{3+} is less than that of Dy^{3+}, so the sublattice magnetization of Pr^{3+} decreases more rapidly than that of Dy^{3+} with temperature increasing,[4] which leads to the minimum magnetic moment μ_s of the alloys between 250 K and 300 K, and hence the minimum value of the magnetostriction in the alloy with $x = 0.6$.

For practical magnetostrictive materials, the magnetostriction in low field is more important than in high field. Therefore, the temperature dependences of the magnetostriction ($\lambda_{||}$) and magnetization (M) of $PrFe_{1.9}$ and $Pr_{0.95}Dy_{0.05}Fe_{1.9}$ were measured during cooling from 200 K to 10 K at a magnetic field of 5 kOe, which are shown in Figs. 4(a) and 4(b), respectively. Two interesting phenomena could be observed in the figures. Firstly, a sharp decrease of the magnetostriction can be evidently observed. As shown in Fig. 4(a), the magnetostriction of $PrFe_{1.9}$ decreases sharply between 30 K and 70 K and the magnetostriction of $Pr_{0.95}Dy_{0.05}Fe_{1.9}$ decreases relatively gradually between 35 K and 120 K. Correspondingly, the sharp decrease of magnetization over the corresponding temperature interval is also evidently detected in Fig. 4(b). This illustrates that a sharp change of the anisotropy may occur in the corresponding temperature range,[3,18,19] which is attributed to the change of the EMD. Then, the abnormal decrease of magnetostriction in Pr-rich alloys shown in Fig. 3 can be well explained.[3] Take $PrFe_{1.9}$ as an example, the EMD of the alloy lies along [100] between 10 K and 30 K. In this temperature range, the magnetostriction decreases monotonously to a minimum at 30 K due to the decrease of the magnetization. However, between 30 K and 70 K, a change of the EMD from [100] to [111] occurs, which leads to the abnormal decrease of the magnetocrystalline anisotropy to a minimum at 70 K, and hence the maximum of magnetostriction at 70 K at a given magnetic field. The changes of the EMD with temperature decreasing were also found in Mn substitute for Fe in $Sm_{0.88}Dy_{0.12}(Fe_{1-x}Co_x)$,[18,19] $Tb_{0.3}Dy_{0.7}Fe_2$,[20] $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Co_x)$,[21] and $Ho_{0.85}Tb_{0.15}(Fe_{1-x}Co_x)$.[22]

Secondly, Dy substitution yields an increased magnetostriction over a long temperature interval. As seen in Fig. 4(a), the magnetostriction of $Pr_{0.95}Dy_{0.05}Fe_{1.9}$ is larger than that of $PrFe_{1.9}$ between 10 K to 50 K. This result indicates that a small amount of Dy substitution can help to enhance the low-field magnetostriction, not merely at 5 K (as discussed in the inset of Fig. 2), but in a long range of temperature. This particular feature makes the alloy a potential candidate for low-field applications at low temperature.

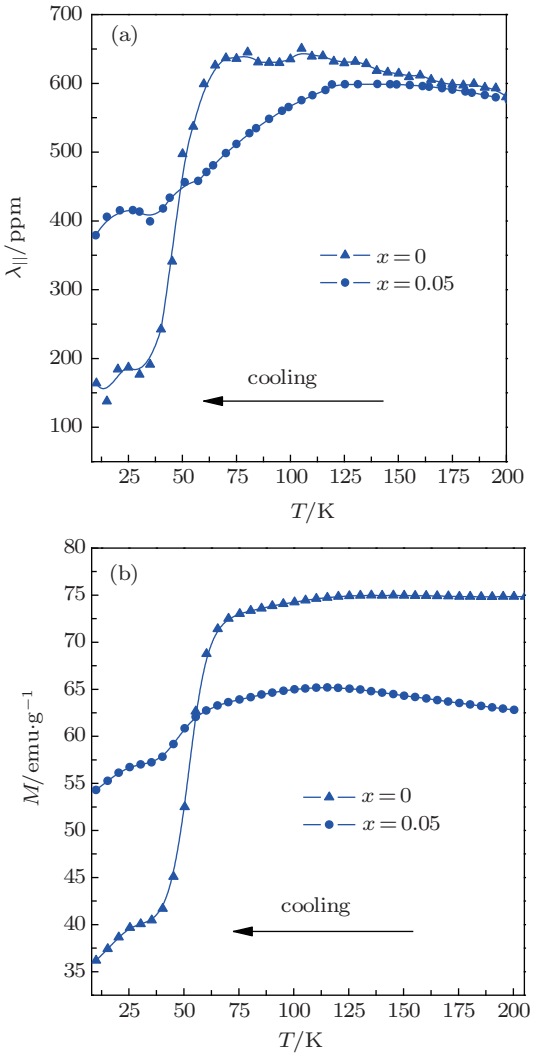


Fig. 4. (color online) The temperature dependences of magnetostriction ($\lambda_{||}$) (a) and magnetization (M) (b) of the alloys with $x = 0.0$ and $x = 0.05$ in a magnetic field of 5 kOe.

4. Conclusion

The temperature dependences of the magnetostriction in polycrystalline $Pr_{1-x}Dy_xFe_{1.9}$ ($0 \leq x \leq 1.0$) alloys between 5 K and 300 K were investigated in detail. Distinct features have been found among the alloys with different Dy contents. For the Dy-rich alloys ($0.4 \leq x \leq 1.0$), the magnetostriction abnormally changes sign at some temperature due to the small and complex nature of λ_{100} of $DyFe_2$. While for the Pr-rich alloys ($0 \leq x \leq 0.2$), the magnetostriction decreases unusually with temperature decreasing. A small amount of Dy substitution ($x = 0.05$) is beneficial to enhance the magnetostriction in low-magnetic field ($H = 5$ kOe) between 10 K and 50 K. These findings have significant impacts and applications in low-temperature and low-magnetic fields.

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