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Composition anisotropy compensation and magnetostriction in $Pr(Fe_{1-x}Co_x)_{1.9}$ ($0 \leq x \leq 0.5$) cubic Laves alloys

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Polycrystalline magnetostrictive alloys $Pr(Fe_{1-x}Co_x)_{1.9}$ ($0 \leq x \leq 0.5$) with cubic Laves phase were synthesized by high-pressure annealing. Measurements of Curie temperature, easy magnetic direction (EMD), and magnetostriction were made on these alloys. The EMD of the alloys rotates continuously from ⟨111⟩ for $x=0.0$ to ⟨110⟩ for $x=0.3$ and then shows a tendency to ⟨111⟩ with further increasing x. Two magnetostriction peaks at low fields are observed around $x=0.2$ and $x=0.4$ due to the lower anisotropy of these two alloys, which is consistent with the variation of EMD. This work demonstrates that the composition anisotropy compensation can be realized in $Pr(Fe_{1-x}Co_x)_{1.9}$ system. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2938705]

Although the compounds RFe_2 (R=rare earth) with C15 cubic Laves structure possess large magnetostriction at room temperature, they are unsuitable as practical materials in magnetostrictive devices because of their high magnetic anisotropy.[1] Therefore, one of the most important aspects in the research of RFe_2 alloys is to reduce the magnetic anisotropy and achieve large values of magnetostriction at low magnetic fields. In the past three decades, much effort has been paid to $R'_xR''_{1-x}Fe_2$ anisotropy compensation system by alloying two different RFe_2 compounds with the same magnetostriction sign but with the opposite signs of their anisotropy constant K_1.[2–5] In these anisotropy compensation systems, the minimum of the magnetic anisotropy frequently occurs when the easy magnetic direction (EMD) changes from ⟨111⟩ to ⟨100⟩ (or ⟨110⟩) at room temperature. It can be inferred that EMD transition is crucial for the realization of good magnetostrictive property in anisotropy compensation systems. For example, in $Tb_xDy_{1-x}Fe_2$ system, the anisotropy compensation is realized at the point of $x=0.27$, which is the critical point of EMD transition from ⟨111⟩ to ⟨100⟩ at room temperature. The similar situation can also be found in other anisotropy compensation systems, such as $Tb_xHo_{1-x}Fe_2$,[1,2] $Tb_{0.2}Dy_{0.8-x}Pr_x(Fe_{0.9}B_{0.1})_{1.93}$,[6,7] and $Tb_xNd_{1-x}Fe_2$.[8]

In analogy with this anisotropy compensation system, we propose that the anisotropy minimum might also be achieved by the appropriate substitution of transition metal in RFe_2 alloys. It is known that the EMD of $PrFe_2$ lies along ⟨111⟩ at room temperature,[9] while that of $PrCo_2$ is ⟨100⟩ below its Curie temperature.[10] Therefore, the composition anisotropy compensation may be also observed around the EMD transition point in $Pr(Fe_{1-x}Co_x)_2$ system which is similar to the $R'_xR''_{1-x}Fe_2$ system.

On the basis of this idea, we attempted to synthesize the $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys. However, $Pr(Fe_{1-x}Co_x)_{1.9}$ with cubic Laves phase cannot be obtained using a traditional high-temperature annealing method when $x<0.6$.[11,12] Recently, high-Pr content magnetostrictive compounds were synthesized using high-pressure synthesis method.[13] In this letter, polycrystalline alloys $Pr(Fe_{1-x}Co_x)_{1.9}$ ($0 \leq x \leq 0.5$) with cubic Laves phase were prepared by this method and the structure, EMD, and magnetostriction of the alloys are investigated.

Ingots with $Pr(Fe_{1-x}Co_x)_{1.9}$ ($x=0.0, 0.1, 0.2, 0.3, 0.4, 0.5$) stoichiometry were prepared by melting the constituent metals in a magnetocontrolled arc furnace in a high-purity argon atmosphere. The as-cast ingots (about 1 g for each sample) were annealed at 6 GPa and 900 °C for 1 h. After that, all the samples were annealed at 300 °C for 48 h in vacuum quartz capsules. Conventional x-ray diffraction (XRD) analysis was carried out using Cu $K\alpha$ radiation with a Rigaku D/Max-gA diffractometer at room temperature. The Curie temperature was detected by a thermal gravitation analyzer with a vertical gradient magnetic field under the samples. The magnetization of the compounds was measured at 300 K using a superconducting quantum interference device magnetometer up to 65 kOe. The ^{57}Fe Mössbauer spectra were collected on a constant accelerated spectrometer with the transmission geometry at room temperature. The source is ^{57}Co in Pd matrix with an activity about 25 mCi. The spectrum was calibrated with a standard α-Fe foil and analyzed by Lorentzian lines in 256 channels using the software MOSSWINN.[14] The linear magnetostriction was measured using standard strain-gauge technique in directions parallel ($\lambda_{\parallel}$) or perpendicular ($\lambda_{\perp}$) to applied magnetic fields at room temperature.

The XRD patterns for $Pr(Fe_{1-x}Co_x)_{1.9}$ with different Co contents are shown in Fig. 1. It can be found that all the alloys mainly consist of cubic Laves phase with $MgCu_2$-type structure, coexisting with minor impurities, i.e., hcp-Pr phase. As we know, $Pr(Fe_{1-x}Co_x)_{1.9}$ ($0 \leq x \leq 0.5$) cannot be synthesized by a high-temperature annealing method at ambient atmosphere.[11,13] It is generally believed that the atom radius plays an important role in the formation of RFe_2 cubic Laves alloys. Owing to the large radius of Pr^{3+}, the cubic Laves phase could not be obtained under normal pressure. Therefore, the formation of the Laves phase should be ascribed to the effect of the high pressure.

The lattice parameters a of the Laves phase in $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys are derived from XRD data and are shown in Fig. 2(a). The lattice parameter decreases from 7.479 Å for $PrFe_{1.9}$ to 7.409 Å for $Pr(Fe_{0.4}Co_{0.5})_{1.9}$ because of the smaller radius of the cobalt ion. The Curie temperatures T_C of Laves phase in $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys are shown

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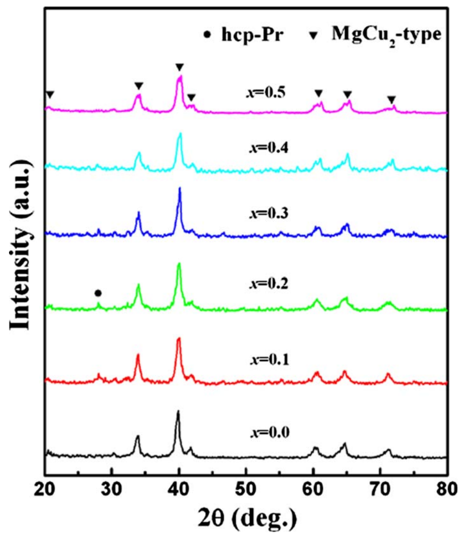

FIG. 1. (Color online) XRD patterns of $Pr(Fe_{1-x}Co_x)_{1.9}$ with different contents of Co.

in Fig. 2(b). The Curie temperature decreases as the Co content increases. It is known that the Curie temperature is mainly determined by the exchange of $3d$-$3d$ atoms in the $R$$Fe_2$ compounds.[15] The coupling between Fe and Co atoms is an essential factor in determining the Curie temperature for the Fe- and Co-containing alloys. Therefore, this result indicates that the magnitude of Co–Co interactions is less stronger than that of the Fe–Fe or Fe–Co interactions in the $Pr(Fe_{1-x}Co_x)_{1.9}$ system.

The room-temperature ^{57}Fe Mössbauer spectra for $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys with $x=0.0, 0.2, 0.3, 0.4$ are shown in Fig. 3(a). The open circles present the experimental data and the lines give the fitted curves. Figure 3(b) gives the Co content dependence of fitted area ratio of two sextets. The data of $PrFe_{1.9}$ can be fitted by two sextets with area ratio about 3:1, which is the characteristic of the EMD along $\langle 111\rangle$.[16] As shown in Fig. 3(b), the area ratio gradually deviates from 3:1 with increasing Co content, which means that the EMD of the alloys is deviating from $\langle 111\rangle$ and shows the characters of nonmajor cubic axis symmetry. When Co content increases to $x=0.3$, the two sextet area ratio is about 1:1, which indicates that the EMD of $Pr(Fe_{0.7}Co_{0.3})_{1.9}$ lies along $\langle 110\rangle$.[16] With further increasing Co content, the EMD deviates from $\langle 110\rangle$ and lies along nonmajor cubic axis. It is thought that the electric-field-gradient tensor might be changed or the symmetry of it will be broken when some

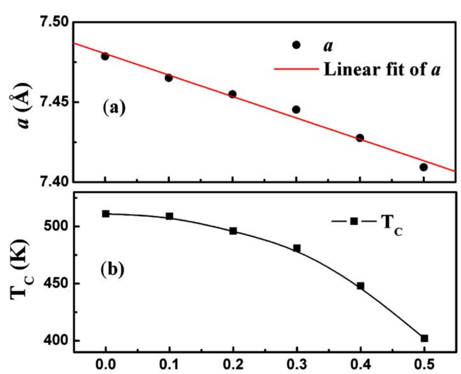

FIG. 2. (Color online) (a) The lattice parameter a and (b) the Curie temperatures T_C of $Pr(Fe_{1-x}Co_x)_{1.9}$ with different contents of Co.

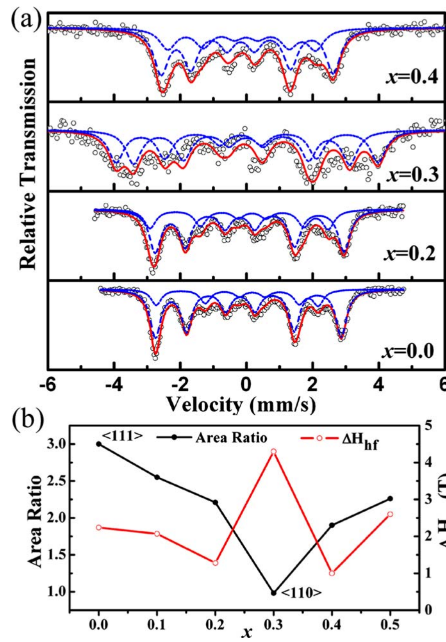

FIG. 3. (Color online) (a) ^{57}Fe Mössbauer spectra at room temperature for $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys. (b) Fitting area ratio and the differences in the hyperfine filed ΔH_{hf} as function of Co content.

Fe sites were occupied by Co in the cubic lattice. As a result, the EMD will change with the substitution Co for Fe in $PrFe_{1.9}$ alloys. The change in EMD with the substitution of transition metal could also be found in some other systems, such as $Tb(Fe_{1-x}Mn_x)_2$,[17] $Tb_{0.27}Dy_{0.73}Fe_{2-x}Ni_x$,[18] $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$,[18] and $Sm_{0.9}Pr_{0.1}(Fe_{1-x}Co_x)$.[19] Figure 3(b) shows the Co content dependence of ΔH_{hf}, where ΔH_{hf} denotes the differences in the hyperfine field H_{hf} between the inequivalent iron sites. It is pointed out that ΔH_{hf} can be attributed to the anisotropic R-Fe exchange interactions in $R$$Fe_2$.[20] We can note that the ΔH_{hf} goes to a maximum near $x=0.3$, which indicates that $Pr(Fe_{0.7}Co_{0.3})$ may have large anisotropy. Two minima can be observed at $x=0.2$ and $x=0.4$, which implies that these two alloys may show low anisotropy.

The magnetization at 300 K of the compounds was measured using a superconducting quantum interference device magnetometer. Figure 4 shows magnetizations curves for $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys at magnetic fields from 0 to 30 kOe. It can be seen that the magnetizations of $Pr(Fe_{0.6}Co_{0.4})_{1.9}$ and $Pr(Fe_{0.8}Co_{0.2})_{1.9}$ are easier saturated, indicating that theses

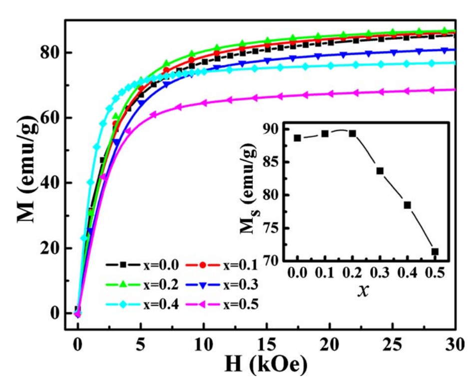

FIG. 4. (Color online) Magnetization curves for $Pr(Fe_{1-x}Co_x)_{1.9}$ at 300 K. The inset shows the saturation magnetization (M_s) with different contents of Co.

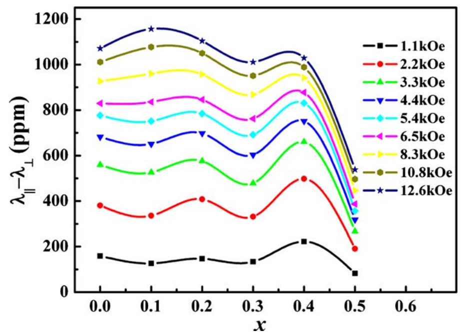

FIG. 5. (Color online) Magnetostriction $\lambda_{\parallel}$-$\lambda_{\perp}$ for $Pr(Fe_{1-x}Co_x)_{1.9}$ at various applied magnetic fields.

two alloys show lower magnetic anisotropy. The saturation magnetization (M_s) derived from M-H curves is shown in the inset of Fig. 4. The saturation magnetization of $Pr(Fe_{1-x}Co_x)_{1.9}$ increases slowly to a maximum around $x=0.2$ and then decreases with increasing Co content. The variations of the saturation magnetization can be understood on the basis of the rigid-band model.[21]

The magnetic field dependence of the magnetostriction ($\lambda_{\parallel}$-$\lambda_{\perp}$) for $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys is shown in Fig. 5. The magnetostriction of $Pr(Fe_{0.7}Co_{0.3})_{1.9}$ is low, ascribed to its EMD along ⟨110⟩ and large magnetic anisotropy. Two peaks at low magnetic fields can be observed around $x=0.2$ and $x=0.4$, due to the low anisotropy of these two alloys. These results are consistent with the above Mössbauer and magnetization analysis. Similar to $R'_xR''_{1-x}Fe_2$ anisotropy compensation system, the magnetostriction peaks can be observed around the EMD transition points in $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys. As a result, the peaks around $x=0.2$ and $x=0.4$ can be described as the anisotropy compensation points in $Pr(Fe_{1-x}Co_x)_{1.9}$ system.

In conclusion, this work presents that the composition anisotropy compensation can be obtained in single rare-earth based $Pr(Fe_{1-x}Co_x)_{1.9}$ alloys. We believe that the findings in the present work might also be used in other RFe_2 system provided that the RFe_2 and RT_2 have the different EMDs similar to $PrFe_2$ and $PrCo_2$ and the EMD transition occurs at room temperature. $Pr(Fe_{0.6}Co_{0.4})_{1.9}$ shows low magnetic anisotropy and large magnetostriction at low magnetic field. Moreover, the alloy only contains inexpensive light rare-earth Pr, which makes it a promising candidate for practical magnetostrictive material.

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