Magnetics, Magetostriction, and Spin Reorientation of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ Alloys with Substitution of Fe by Al

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***Abstract*—The effects of Al substitution for Fe on the structure, magnetism, magnetostriction, anisotropy and spin reorientation of a series of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloys (x=0. 05，0.1，0.15，0.2，0.25，0.3) have been investigated. The alloys of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ substantially retain $MgCu_2$-type C-15 cubic Laves phase structure when x<0.2. The mixed phases appear with x=0.2, and cubic Laves phase decreases with increasing x. The measurements of magnetics indicate that anisotropy decreases and EMD deviates from the major axis with temperature and composite. The magnetization decrease gradually with increasing temperature, and increases slightly with increasing of Al content x and then decreases rapidly when x>0.15. Coercivcity decreases rapidly with increasing of temperature, and appears the maximum value at Al content x=0.20. The magnetostriction of the $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloys decreases drastically with increasing x and the giant magnetostrictive effect disappears for x>0.15. Fortunately, a small amount of Al substitution is beneficial to a decrease in the anisotropy. The spin reorientation temperature decreases with increasing x. The magnetic properties are determined by both Pr and Al substitution effects.**

Keywords-Cubic laves phase; Magnetostriction; Spin reorientation; EMD

I. Introduction

The pseudobinary rare-earth iron alloy $Tb_{0.3}Dy_{0.7}Fe_2$ (commercially known as Terfenol-D) as an excellent magnetostrictive material can be used as ultrasonic transducers and micro-actuators with giant magnetostrition and relatively low magnetocrystalline anisotropy [1]. However, its application is somewhat limited because of its low tolerance to tensile and shear forces, low electrical resistivity and relatively high saturation field. Many research works have focused on substituting Fe with other elements in an attempt to improve the magnetic and magnetostrictive properties in the $Tb_{0.3}Dy_{0.7}(Fe_{1-x}T_x)_2$ (T= Mn, Co, Ni, Ga, Al, B, Be, etc.) alloys[2-5] and also to improve its application properties. Judging by these research reports, Al is regarded as an ideal substitution for Fe. The application properties of $Tb_{0.3}Dy_{0.7}(Fe_{0.9}Al_{0.1})_{1.95}$ alloys turn out excellent[5-11] because the addition of Al to Fe increases resistivity and ductility and decreases anisotropy, but its saturated magnetostriction greatly decreases. On the basis of retaining the good application properties, we anticipate that substitution of other elements for Tb or Dy enhances the saturated magnetostriction of $Tb_{0.3}Dy_{0.7}(Fe_{0.9}Al_{0.1})_{1.95}$ alloys.

Pr is chosen as an ideal substitute for Fe among the numerous rare earth elements because the single ion model [1, 12] indicates that the magnetostriction of $PrFe_2$ (λ_{111}=5600ppm) is larger than that of $TbFe_2$ (λ_{111}=4400ppm), $DyFe_2$ (λ_{111}=4200ppm) and $SmFe_2$ (λ_{111}=-3200ppm) at 0K. Furthermore, the sign of magnetostriction of $TbFe_2$, $DyFe_2$ and $PrFe_2$ is the same, but the sign of anisotropy of $TbFe_2$ is reversible with that of $DyFe_2$ and $PrFe_2$. Clark et al [10] investigated the magnetocrystalline anisotropy of RFe_2 series with the results that the compensating multi-element composition of RFe_2 (R is the two or three elements) retains large magnetostriction and decreasing magnetocrystalline anisotropic, which is composed of these RFe_2 with the same magnetostriction sign and opposite magnetocrystalline anisotropy. Lou [7] and Zhan [8] groups investigated the effect of Pr substitution for Dy on the anisotropy, magnetostriction and magnetism of the various RFe_2 alloys. Their results show that Pr substitution decreases the anisotropy and Curie temperature, and makes the change of magnetostriction complicated.

Up to now, the research works about the simultaneous substitution effect of rare earth sublattice and transition metal sublattice have been rather rare. In this paper, by substituting Pr for Dy and Al for Fe, we have investigated Al content dependence of crystal structure, magnetization, coercivity, anisotropy, magnetostriction and spin reorientation of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloys. Thus a kind of excellent and potential giant magnetostrictive materials would be developed.

II. Experimental Details

All of the $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ (x=0. 05, 0.1, 0.15, 0.2, 0.25, 0.3) alloys were prepared by melting an appropriate amount of constituent metals in an arc-melting furnace under a high purity argon atmosphere and by remelting several times to achieve good homogeneity. The purity of Tb, Dy and Pr was 99.98%, and that of Fe and Al was 99.99%. An excess of 4wt% Dy was added to compensate for evaporation losses. The button-shaped samples were wrapped in Ta foil, sealed in evacuated quartz tubes filled with high purity argon, homogenized at 950°C for at least 50 hours and then cooled in the air.

X-ray diffraction analysis was performed by means of a Rigaku D/max-2400 diffractometer with a pyrolytic graphite monochromator using Cu-Kα radiation ($\lambda \approx 1.5405$ Å). In the applied field of 900kA/m, with vibrating sample magnetometer (VSM), we measured respectively the magnetization M at room temperature, saturated magnetization σ_s, coercivity H_c for $Tb_{0.3}Dy_{0.7-x}Pr_x(Fe_{0.9}Al_{0.1})_{1.95}$ alloys.

Magnetostriction measurements and relative susceptibility were in turn taken by a standard strain gauge in applied fields up to 800kA/m at room temperature and multifunction magnetic measurement system WDCCC at 100-380K. The samples, which were cut from the button by a linear spark erosion cutter, were typically 5mm $\times$ 6mm $\times$ 2mm.

III. RESULTS AND DISCUSSIONS

The powder X-ray diffraction patterns of the $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ (x=0. 05, 0.1, 0.15, 0.2, 0.25, 0.3) alloys is shown in Fig1 indicating that the homogenized alloys substantially retain $MgCu_2$-type C-15 cubic Laves phase structure when x<0.2. The mixed phases appear with x=0.2 and increase with increasing x.

In the applied field of 900kA/m, with vibrating sample magnetometer (VSM), we measured respectively the magnetization M at room temperature, coercivity H_c, saturated magnetization σ_s for $Tb_{0.3}Dy_{0.7-x}Pr_x(Fe_{0.9}Al_{0.1})_{1.95}$ alloys. Fig.2 shows the room temperature magnetization curve of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloys at various AI content x. Obviously, it is shown that the change of magnetization of the alloys is complicated with increasing Al content x. Substituting of Dy by Pr increases the exchange-coupling interaction between Fe and Fe, as a result, the magnetization of the alloys decreases. Simultaneously substituting of Fe by a small amount of Al ($x \le 0.1$) reduces the number of Fe atom couple, weakening the entire exchange-coupling interaction between Fe atoms so that the magnetization of the transition metal sublattice decreases, and therefore the magnetization of the alloys increases. Continuing to add more Al, the number of Fe atoms decreases further. Consequently, not only weakens the exchange coupling between Fe-Fe and affects the exchange interaction between R-Fe and R-R, but also the magnetization of transition-metal and rare-earth metal sublattice reduces in varying degrees. Pr and Al substitution effect determines together the magnetization of the alloys and makes the behaviour of magnetization complicated. Al substitution effect completely destroys the spin exchange coupling between Fe-Fe, R-Fe and R-R when x>0.25 so that $Tb_{0.3}Dy_{0.7-x}Pr_x (Fe_{0.9}Al_{0.1})_{1.95}$ alloys are of paramagnetism, in which magnetization increases linearly with the external magnetic field. Comparing to the results in our previous investigations[4, 10], we found that Pr substitution improve the Al content of the ferromagnetic alloys. Similarly, Fig.2 indicates that the magnetization is saturated more easily, implying magnetocrystalline anisotropy reduces in the $Tb_{0.3}Dy_{0.7-x}Pr_x(Fe_{0.9}Al_{0.1})_{1.95}$ alloys.

Temperature dependences of coercivity H_c and saturated magnetization σ_s of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloy is shown in Fig.3. It is found that the alloys possess the characteristics of low temperature hard magnetism[13] and the coercivity experiences drastic changes with temperature and composition, indicating EMD or spin reorientation rotates and deviates from the major axis [111] in the plane {110} with temperature and Al substitution, and thus the magnetostriction correspondingly decreases. Coercivcity decreases rapidly with increasing of temperature, and appears the maximum value at Al content x=0.20. Form Fig.3, we also see that saturated magnetization decrease gradually with increasing temperature, and increases slightly with the increase of Al substitution and then decreases rapidly when substitute x>0.15.

The magnetic field dependence of the magnetostriction λs for $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ (x=0. 05, 0.1, 0.15, 0.2) alloys is shown in Fig.4 where the magnetostriction λs increases with the applied field, but it decreases because of substituting Fe by Al. Furthermore, the magnetostriction of the Al substituted samples is more easily saturated in an applied field with the increase of x, implying that the substitution of Al might be beneficial to decreasing the magnetocrystalline anisotropy and thus leads to the transformation of EMD, i.e. the spin reorientation, which is consistent with the results of VSM measurements. The decrease of anisotropy and the change of EMD result from the Al substitution for Fe. From Fig.4, it is observed that the magnetostriction λs is merely 440×10^{-6} for x=0.15, namely, it equals 60% of x=0. 05 and magnetostrictive effect vanishes for x>0.15. λs is less than 130×10^{-6}.

We investigated Al content dependence of spin reorientation temperature Tm of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ (x=0. 05, 0.1, 0.15, 0.2, 0.25, 0.3) alloys, as shown in Fig.5. It is observed that the spin reorientation temperature decreases rapidly with increasing Al content x. Because the substitution of Al for Fe decreases anisotropy, the spin reorientation happens at a low temperature. Simultaneously, the spin reorientation temperature T_m of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ is found larger than that of $Tb_{0.3}Dy_{0.6}(Fe_{1-x}Al_x)_{1.95}$ due to substitution of Dy by Pr. The result of spin reorientation temperature T_m is in good agreement with the results of magnetism and magnetostriction measurements.

IV. CONCLUSION

The investigations of x-ray diffraction, magnetics, magnetostriction, spin reorientation for $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ (x=0. 05, 0.1, 0.15 0.2, 0.25 0.3) alloys indicate that the alloys substantially retain $MgCu_2$-type C-15 cubic Laves phase structure when x<0.2 and the mixed phases appear with x=0.2 and increase with increasing x. The magnetic properties are determined by both Pr and Al substitution effects. The measurements of magnetization, coerecivity and magetostriction indicate that magnetocrystalline anisotropy decreases and EMD deviates from the major axis with temperature and Al content x. The magnetization decrease gradually with increasing temperature, and increases slightly with increasing of Al content x and then decreases rapidly when x>0.15. Coercivcity decreases rapidly with increasing of temperature, and appears the maximum value at Al content x=0.20. The magnetostriction decreases drastically with increasing x and the giant magnetostrictive effect disappears for x>0.15, but a small amount of Al substitution is beneficial to a decrease in the anisotropy. The spin reorientation temperature decreases with increasing x.

ACKNOWLEDGEMENTS

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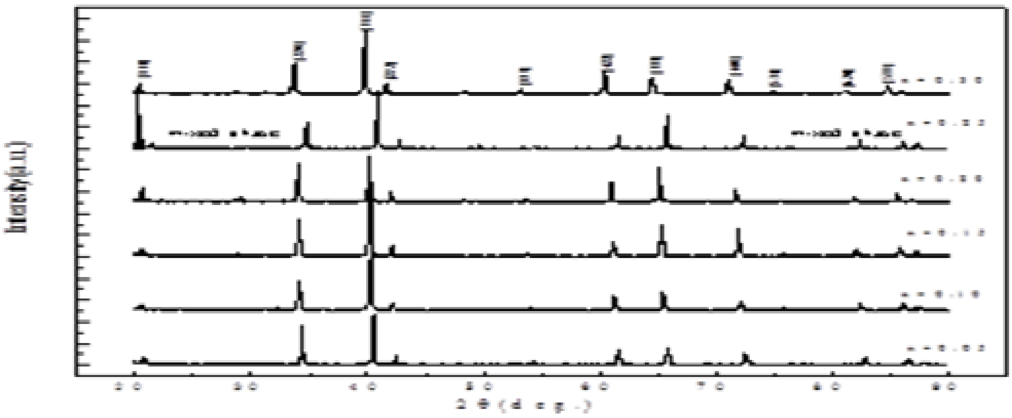

Figure 1. X-ray diffraction patterns for the $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloys

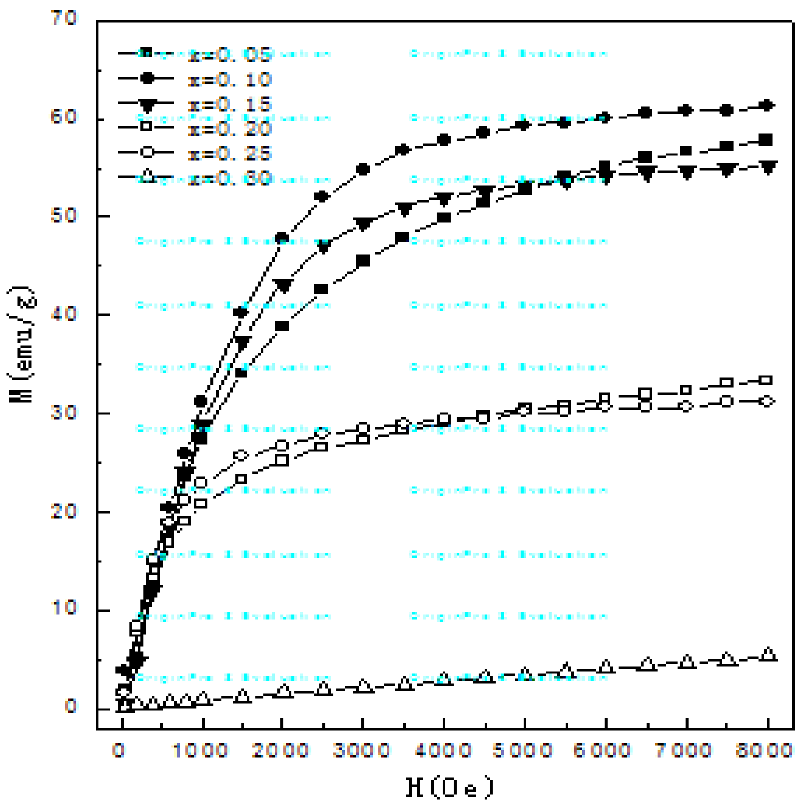

Figure 2 Magnetization curve of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloys at room temperature

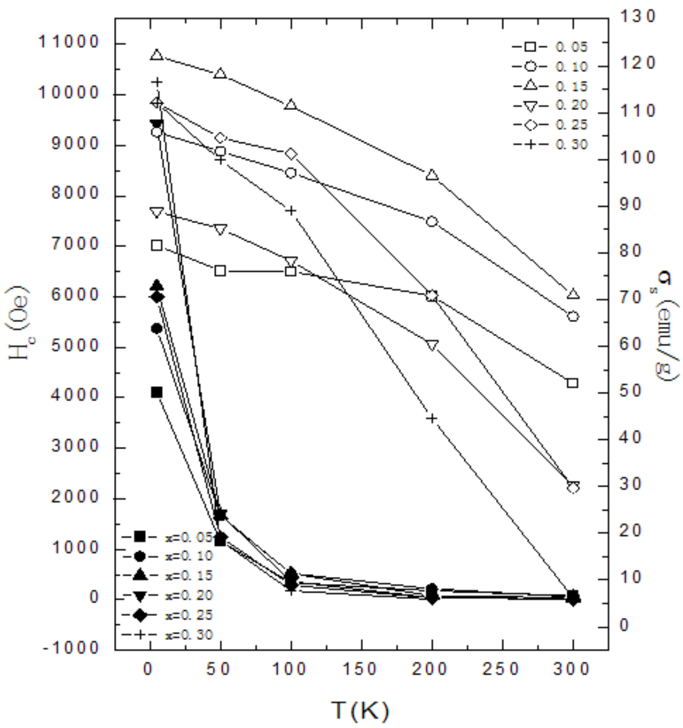

Figure 3. Temperature dependences of coercivity and saturated magnetization of $Tb_{0.3}Dy_{0.6}Pr_{0.1}(Fe_{1-x}Al_x)_{1.95}$ alloys

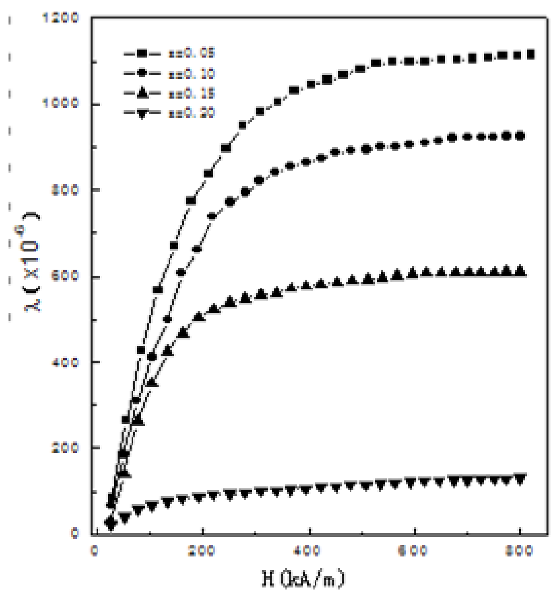

Figure 4. Magnetostriction λ for $Tb_{0.3}Dy_{0.6}Pr_{0.1}$ (Fe1-xAlx)$_{1.95}$ alloys as a function of the applied field

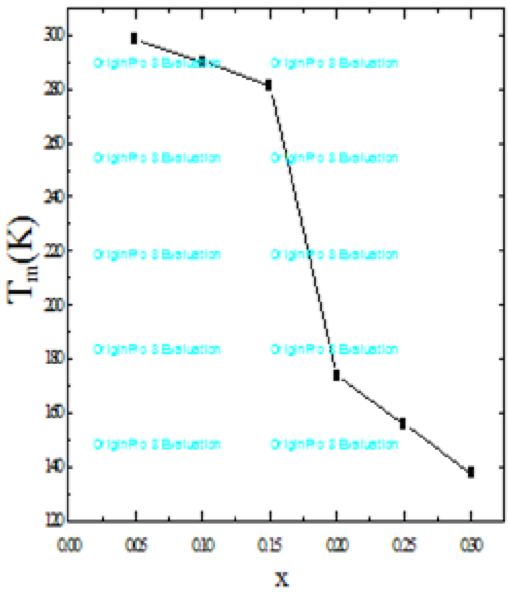

Figure 5. Al content dependence of spin reorientation temperature T_m of $Tb_{0.3}Dy_{0.6}Pr_{0.1}$ $(Fe1\text{-}xAlx)_{1.95}$ alloys