# Anisotropy and FOMP in $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0–1)$ and $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0–0.4)$ Compounds

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***Abstract*—Alloys of composition $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}$ $(x = 0–1)$ and $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}$ $(y = 0–0.4)$ were synthesized and characterized in the temperature range of 10–1273 K in fields up to 5 T. The magnetization curves along the hard direction clearly indicate the presence of a Type II FOMP (first order magnetization processes) below 200 K. The critical fields $H_{cr}$ at which FOMP is observed, have been determined from the occurrence of a maximum in $dM/dH$ as a function of $H$. For $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}$ compounds, $H_{cr}$ is the largest (4.4 T at 5–10 K) for the $x = 0$ composition, and the lowest (1.4 T at 5–10 K) for the $x = 0.5$ composition. For the $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}$ compounds, the $H_{cr}$ are about 3.6 T at 5–10 K for $y = 0$ and 0.1 whereas, $H_{cr}$ is seen to decrease monotonically for higher Co contents. The FOMP exhibits a strong monotonically decreasing temperature dependence for both systems. The effects of the R sublattice (Sm and Pr) and the 3d sublattice (Fe and Co) on the anisotropy and FOMP are reported.**

***Index Terms*—Alloys, anisotropy, FOMP, Sm–Pr–Fe–Co–Ti.**

## I. Introduction

THE $R_3(Fe, M)_{29}$ $(3 : 29)$ compounds, which have been reported as potential candidates for high temperature permanent magnet applications, crystallize in monoclinic structure with $A2/m$ space group. This structure is formed by alternative stacking $1 : 12$ and $2 : 17$ type segments. There are two crystallographically inequivalent sites for the rare earth occupancy viz., 2a and 4i sites [1]. The 2a site has a local $1 : 12$ like environment and the 4i site has a $2 : 17$ like environment [2]. The two R sites have opposite signs of $A_{20}$ (namely, $A_{20} < 0$ for the 4i site and $A_{20} > 0$ for the 2a site). Thus, the competition of the two R sublattices may result in an unusual magnetization behavior. In fact, the type II first-order magnetization processes (FOMP) have been observed in the compounds of $Sm_3(Fe, Ti)_{29}$ [3], $Nd_3(Fe, Ti)_{29}$ [4] and $Pr_3(Fe, Ti)_{29}$ [5], [6]. In the present work, these studied have been extended to quarternary-alloy systems with two different R atomic species or two different 3d atoms. Alloys of composition $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0–1)$ and $Pr_3(Fe_{1-x}Co_x)_{27.5}Ti_{1.5}(y = 0–0.4)$ were synthesized and characterized in the temperature range of 10–1073 K in fields up to 5 T. The effects of the R sublattice (Sm and Pr) and the 3d sublattice (Fe and Co) on the anisotropy and FOMP are reported.

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## II. Experimental Detail

Alloys of composition $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0–1)$ and $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0–0.4)$ were prepared by arc melting under argon atmosphere and then annealed at 1000–1200 K. Powder X-ray diffraction (XRD) with Fe $K_\alpha$ radiation and thermomagnetic analysis (TMA) were used to determine the crystal structure, the phases present, the unit cell parameters, and their Curie temperatures. Magnetic properties were measured in the temperature range between 5 K and 300 K in fields up to 5T using a superconducting quantum interference device (SQUID) magnetometer. The magnetically oriented powder samples were prepared by mixing fine powders ($\leq 20$ $\mu$m) with epoxy and aligning in a field of 2.5 T. Magnetization measurements have been carried out on the oriented powers, along and perpendicular to the alignment direction. The FOMP can be only observed in magnetization curves taken with the field oriented along the hard (perpendicular) direction. The critical fields, $H_{cr}$, at which FOMP is observed, have been determined from the maximum in the $dM/dH$ versus $H$ curves. The anisotropy field ($H_A$) was estimated by extrapolation of the difference between the easy axis $M$ and hard axis $M$ to zero.

## III. Results and Discussions

### A. $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0–1)$

XRD data taken for random $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0–1)$ samples indicated that all alloys have monoclinic structures. The XRD patterns for aligned (perpendicular) samples are shown in Fig. 1. XRD for magnetically aligned samples allows for the determination of the easy magnetization direction (EMD). For $x = 0.2$ ($Sm_{0.2}$ $Pr_{0.8}$), a strong reflection corresponding to (040), which is the same as that in $Pr_3Fe_{27.5}Ti_{1.5}$ [6], is observed. This implies that the EMD is near the $b$ axis. However, with increasing of Sm content, the strongest reflections become the (204) and (402) for $x = 0.5$ and (402) for $x = 0.8$, respectively. This indicates that the EMD has changed to the

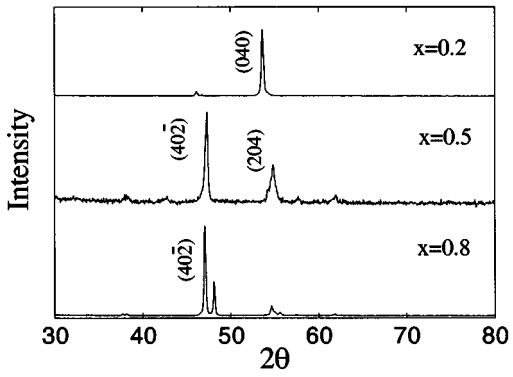


Fig. 1. XRD of $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x{=}0{-}1)$ in aligned (perpendicular) powder sample.

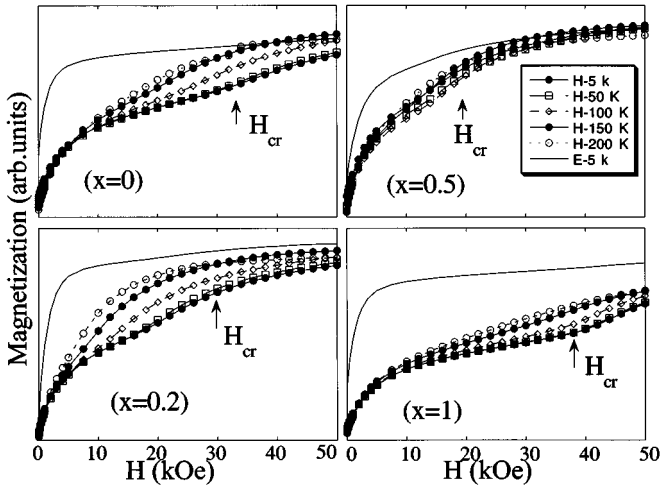


Fig. 2. $M$ versus $H$ of $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x{=}0\text{-}1)$. ($E$–$M \| H$, $H$–$M \perp H$).

ac-plane, which is similar to that observed in $Sm_3Fe_{27.5}Ti_{1.5}$ [3]. This also indicates that the alloy with $y = 0.5$ shows the weakest anisotropy.

Magnetization curves for the $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0\text{–}1)$ compounds, with the field aligned along the easy and the hard directions, respectively, are shown at various temperatures in Fig. 2. A discontinuity of the magnetization curve along the hard magnetization has been observed at below 150–200 K for all compounds. This phenomenon is the signature of a FOMP, which is an irreversible rotation of the magnetization vector $M_s$ under the action of an applied magnetic field, which results in a discontinuity of the magnetization curve along the hard magnetization direction [7]. Since the $M_s$ jump does not achieve saturation, it is classified as a Type II FOMP. The critical field, $H_{cr}$, at which the FOMP is observed, can be determined from maximum in the $dM/dH$ versus $H$ curves. These have been plotted in Fig. 3.

The FOMP shows a strong composition and temperature dependence as shown in Fig. 4(a). The $H_{cr}$ is the largest (~4.4 T at 5–10 K) for the $x = 0$ (Sm) and the lowest (~$1.4T$ at 5–10 K) for the $x = 0.5$ ($Sm_{0.5}Pr_{0.5}$) sample. For $x = 0.2$ and 0.8, $H_{cr}$ at 5–10 K are ~2.1 T and ~3.6 T, respectively. For $x = 0$ (Pr) sample, the $H_{cr}$ is ~3.6 T. The $H_{cr}$ for all compounds

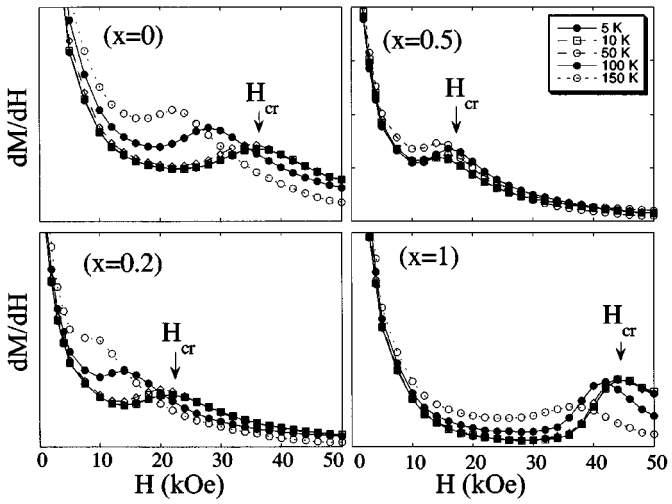


Fig. 3. $dM/dH$ versus $H$ for $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0\text{-}1)$. (Taken from $M$ versus $H$ along the hard direction.)

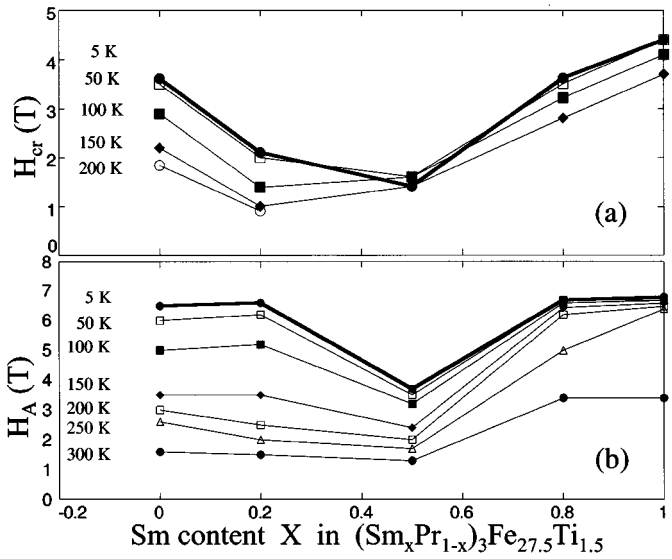


Fig. 4. $H_{cr}$ and $H_A$ versus $x$ of $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0\text{–}1)$. (a) $H_{cr}$ versus $x$, (b) $H_A$ versus $x$.

are monotonically decreasing and the FOMP becomes less pronounced with increasing temperature. The FOMP nearly disappears at about 150–200 K. The anisotropy fields $H_A$ also show a strong composition and temperature dependence, which is similar to that of the $H_{cr}$, As seen in Fig. 4(b), the $y = 0.5$ sample also shows the lowest $H_A$ value. This result is consistent with the XRD measurements on the aligned samples (Fig. 1).

As mentioned above, the above unusual magnetization behavior may be attributed to the competition of the two R sublattices with opposite signs of $A_{20}$, and also the competition of two different R atoms occupying two different R sites, 2a and 4i. The lower $H_{cr}$ or $H_A$ at $x = 0.5$ may be attributed to a strong cancellation between the different types of anisotropies contributed by the two R sublattices. Our earlier unpublished works, the magnetization, Mossbauer and ac susceptibility measurements [8], indicated that the Sm atoms may prefer to occupy the 2a sites and the Pr atoms may prefer to occupy the 4i sites. We believed that the above magnetization behavior can

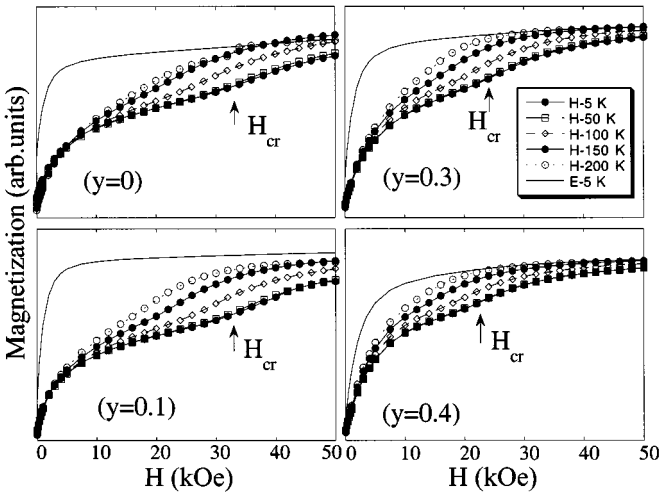


Fig. 5. $M$ versus $H$ of $(Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y{=}0\text{–}0.4)$. $(E\text{–}M \| H,\ H\text{–}M \perp H)$.

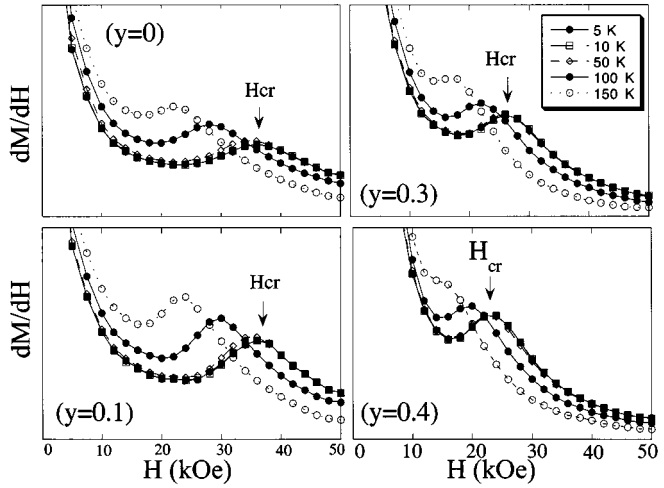


Fig. 6. $dM/dH$ versus $H$ of $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0\text{–}0.4)$. (Taken from $M$ versus $H$ along the hard direction.)

be also related to the R atom sites occupation preference. In order to understand the above complex magnetic behavior. A first-principles theoretical model calculation, incorporating exchange and crystal-field interaction, is in progress. These calculation results will be published soon.

### *B. $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0\text{-}0.4)$*

Using the method described above, we have also observed the composition and temperature dependence of the FOMP and the anisotropy in the $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0\text{-}0.4)$ compounds (Figs. 5–7). As shown in Fig. 7(b), the $H_A$ for the $y = 0.1$ composition is slightly higher than that of $y = 0$ composition in the temperature range between 5 K and 300 K. It then decreases monotonically for higher Co contents. These results are consistent with our prior results [6], in which the XRD of magnetically oriented samples shows that the EMD is along

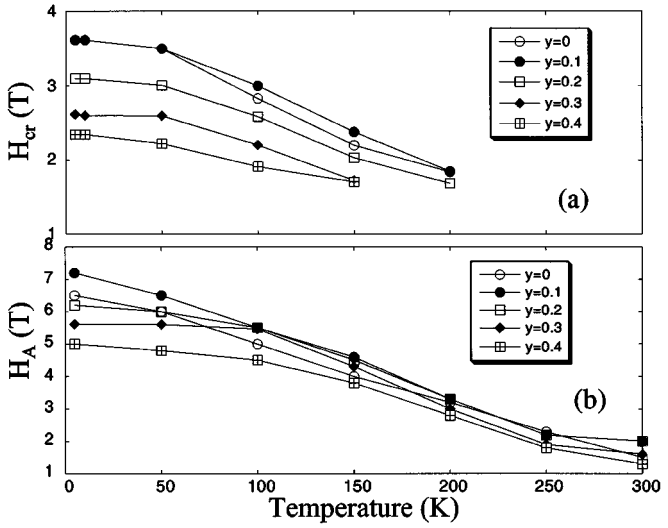


Fig. 7. $H_{cr}$ and $H_A$ versus T of $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0\text{–}0.4)$. (a) $H_{cr}$ versus T, (b) $H_A$ versus T.

the $b$-axis for $y(\text{Co}) = 0.1$, and nearly along the $b$-axis for the $y = 0$, 0.2, and 0.3 compositions. The anisotropy behavior of the 3d(Fe) sublattice has also been changed by Co substitution for Fe in the $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0\text{–}0.4)$. $H_{cr}$ shows a strong composition and temperature dependence, which is similar to that of the $H_A$. As shown Figs. 5 and 6, the FOMP becomes less pronounced with increasing temperature and disappears at above 150 K. The values of $H_{cr}$ and $H_A$ decreases with increasing temperature.

## IV. Conclusion

The type II FOMP have been observed in the $(Sm_xPr_{1-x})_3Fe_{27.5}Ti_{1.5}(x = 0\text{-}1)$ and the $Pr_3(Fe_{1-y}Co_y)_{27.5}Ti_{1.5}(y = 0\text{–}0.4)$ compounds below 200 K. Both the FOMP and anisotropy fields exhibit a strong temperature and composition dependence for these two systems. A theoretical analysis regarding the above composition and temperature dependence of the FOMP and anisotropy field is in progress.

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