STRUCTURAL AND MAGNETIC PROPERTIES OF $(Pr_xSm_{1-x})Mn_2Si_2$ ①

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ABSTRACT Crystallographic and magnetic properties of intermetallic compounds $(Pr_xSm_{1-x})Mn_2Si_2$ ($x = 0 \sim 0.80$) have been investigated by X-ray powder diffraction, XPS and magnetic measurements. All the compounds crystallize in $ThCr_2Si_2$-type structure. Substitution of Pr for Sm leads to the increase of the lattice constants and the transition from antiferromagnetism (AFM) to ferromagnetism (FM). The valence-fluctuation in the compounds was observed and the relation between the change of electron binding energy and magnetic properties was also discussed preliminarily.

Key words rare earth intermetallic compounds magnetic materials

1 INTRODUCTION

Ternary layered intermetallic compounds RMn_2X_2 (R = rare earth and X = Si, Ge) have become of renewed interest recently because of the large magnetoresistance (MR) effect[1-3] which quite resembles that of the artificially layered magnetic thin film[2,4]. The low temperature magnetic phase transitions and magnetoresistive effects of $SmMn_2Si_2$ have been investigated in our previous work, there exist a ferromagnetic transition at 35 K and two antiferromagnetic transitions at 120 K and 230 K[5]. The structure and magnetic properties of the $(Nd_xSm_{1-x})Mn_2Si_2$ are also studied[6].

The substitution of Pr for Sm aimed at not only improving the performance of the magnetic materials, but also examining the magnetic interaction between the magnetic moments of rare earth and transition metal. This paper is one of our series works on structure and magnetic properties of $(R_xR'_{1-x})Mn_2Si_2$ and deals with the influence of substitution of Pr for Sm on the crystal structure, magnetic properties and valence-fluctuation in $(Pr_xSm_{1-x})Mn_2Si_2$.

2 EXPERIMENTAL

The samples were prepared by arc-melting of stoichiometric mixture of starting materials (purity > 99.9 % for Pr, Sm, Mn and purity > 99.99 % for Si) in Ar atmosphere. The X-ray powder diffraction analysis and Magnetic measurements were carried out as in Ref.[6].

3 RESULTS AND DISCUSSION

3.1 Crystallographic data of $(Pr_xSm_{1-x})Mn_2Si_2$

X-ray powder diffraction analysis indicated that all the synthesized samples were single phase with body-centered tetragonal $ThCr_2Si_2$ structure. The XRD pattern of $(Pr_xSm_{1-x})Mn_2Si_2$ ($x = 0.35$) is shown in Fig.1. The lattice parameters are given in Table 1. It is found that substitution of Pr for Sm leads to an increase of the lattice constants a and c with increasing Pr content x. This associates with the larger atomic radius of Pr (1.828 Å) compared with Sm

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(1.802 Å). At the same time, the intralayer Mn-Mn distance R^a(Mn-Mn) and the interlayer Mn-Mn distance R^e(Mn-Mn) should be changed.

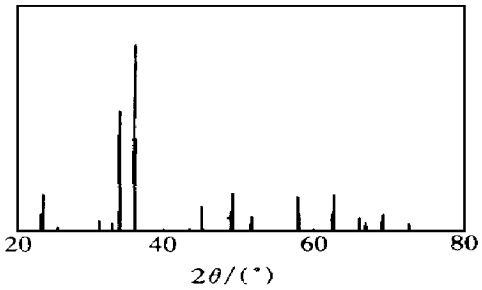

Fig.1 XRD pattern of $(Pr_xSm_{1-x})Mn_2Si_2$

3.2 Magnetic properties of $(Pr_xSm_{1-x})Mn_2Si_2$

Magnetic hysteresis loops of the compounds were measured by vibrating sample magnetometer (VSM) at room temperature in a field of 1 T. Coercivity H_c, retentivity B_r and saturation magnetization M_s are given in Table 2, which shows that $SmMn_2Si_2$ is antiferromagnetic with $T_N = 398$ K[7], partial substitution of Pr for Sm leads to the transition from AFM to FM, and when Sm is fully replaced by Pr, the resulting compound $PrMn_2Si_2$ turns into antiferromagnetic again with $T_N = 348$ K[7]. This change from AFM to FM, then FM to AFM may be resulted from the change of R^a(Mn-Mn) after substitution of Pr for Sm, and the influence of Sm-Mn, Pr-Mn exchange interactions[8].

Table 1 Crystallographic data of $(Pr_xSm_{1-x})Mn_2Si_2$

x	a/Å	c/Å	V/Å^3	c/a
0.00	3.9750	10.517	166	2.6457
0.10	3.9923	10.542	168	2.6406
0.35	4.0049	10.552	169	2.6348
0.80	4.0240	10.560	171	2.6243
1.00*	4.0250	10.555	171	2.6220

* —Rossi D *et al*. J Less—Common Met, 1978, 58: 203.

Partial substitution of Pr for Sm leads to the transition from weak magnetism to strong magnetism, H_c of $(Pr_{0.80}Sm_{0.20})Mn_2Si_2$ is up to 2.448 kOer, and B_r, M_s is 3.268 emu/g and 4.902 emu/g, respectively. H_c, B_r and M_s of the compounds are closely related to the Pr concentration, they increase with increasing Pr content x. Fig.2 shows the magnetization curves of the compounds for $x = 0.10$, $x = 0.35$ and $x = 0.80$ in the temperature from 80 to 298 K, it is thus clear that there is no magnetic transition in this temperature range.

Table 2 Magnetic data of $(Pr_xSm_{1-x})Mn_2Si_2$ series

x	H_c/Oer	B_r/(emu·g^{-1})	M_s/(emu·g^{-1})
0.00	AF(T_N = 398 K)		
0.10	150.0	0	0.7973
0.35	947.2	0.1592	0.7962
0.80	2448.0	3.2680	4.9020
1.00	AF(T_N = 348 K)		

3.3 Valence fluctuation in compounds

Binding energy of the elements in the compounds are listed in Table 3. Compared with the binding energy in single element state, the binding energy of Sm_{3d5}, Pr_{3d} and Mn_{2p} in the compounds increases while that of Si_{2p} decreases, especially the change of binding energy of Sm_{3d5} increases to 2.0 eV. It is implied that the electron cloud densities of Sm_{3d5}, Pr_{3d} and Mn_{2p} decrease and the valences increase at the same time while that of Si_{2p} increases and the valence de-

Table 3 Electron binding energy(eV) of elements*

	Sm_{3d5}	Δ	Pr_{3d}	Δ
Single element state	1082.7		932.5	
$(Pr_{0.80}Sm_{0.20})Mn_2Si_2$	1084.7	2.0	933.1	0.6
	Mn_{2p}	Δ	Si_{2p}	Δ
Single element state	640.7		98.8	
$(Pr_{0.80}Sm_{0.20})Mn_2Si_2$	641.1	0.4	98.2	-0.6

* —The measurement error for XPS is 0.2 eV.

creases at the same time. So we think that there is valence fluctuation in the compounds as observed in $SmMn_2Si_2$[9] and in (Nd_xSm_{1-x})-Mn_2Si_2[6]. It seems that there is some relation between the change of magnetic properties and the change of binding energy, and further studies are in progress.

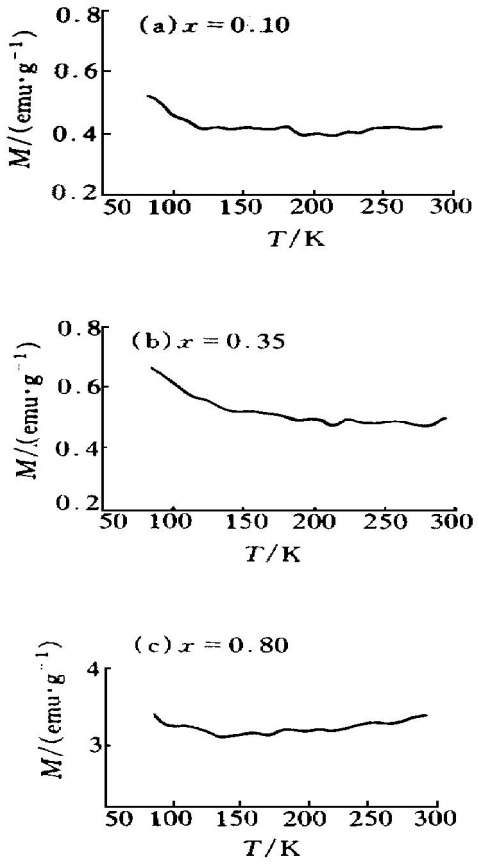

Fig. 2 Magnetization curves of (Pr_xSm_{1-x}) Mn_2Si_2

4 CONCLUSIONS

(1) All the compounds crystallize in $ThCr_2Si_2$-type structure. Substitution of Pr for Sm leads to an increase of the lattice constants a and c with increasing Pr content x. This associates with the larger atomic radius of Pr(1.828 Å) compared with Sm (1.802 Å).

(2) Partial substitution of Pr for Sm leads to the transition from weak magnetism to strong magnetism, which may be resulted from the change of R^a(Mn-Mn) after substitution of Pr for Sm, and the influence of Sm-Mn, Pr-Mn exchange interactions.

(3) Compared with the binding energies in single element state, the binding energies of Sm_{3d5}, Pr_{3d} and Mn_{2p} increase while that of Si_{2p} decreases. It is implied that there is valence-fluctuation in (Pr_xSm_{1-x}) Mn_2Si_2.

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